

### 3.6.8 Uranium and Enriched Uranium Focus Area in Pit 5

Three areas in Pit 5 were investigated to locate areas of enriched uranium and other uranium waste streams. One area in the northern part of Pit 5 was selected based on monitoring data. The other two areas were selected using WasteOScope. Probe names and location and completion intervals for probes installed in this investigation are provided in Table 3-33 and illustrated in Figure 3-40. The individual areas probed in Pit 5 are discussed in the following subsections.

Table 3-33. Clusters of probes supporting uranium waste assessment in Pit 5.

Cluster Name	Probe Type					
	Type A Probe		Soil-Moisture Probe		Lysimeter	
	Probe Name	Probe Depth (ft)	Probe Name	Instrument Depth (ft)	Probe Name	Port Depth (ft)
<b>Pit 5-1</b>						
	Pit 5-1-1	7.8	—	—	—	—
	Pit 5-1-2	7.8	—	—	—	—
	Pit 5-1-3	9.1	—	—	—	—
	Pit 5-1-4	8.5	—	—	—	—
	Pit 5-1-5	3.8	—	—	—	—
	Pit 5-1-6	11.7	—	—	—	—
	Pit 5-1-7	16.0	—	—	—	—
	Pit 5-1-8	13.6	—	—	—	—
<b>Pit 5-4</b>						
	Pit 5-4-1	16.5	Pit 5-4-M	10.2	Pit 5-4-L1	10.6
	Pit 5-4-2	16.4	Pit 5-4-MB	8.2	Pit 5-4-L3	13.1
	Pit 5-4-3	16.3	Pit 5-4-MB	2.8	—	—
	Pit 5-4-4	12.7	—	—	—	—
	Pit 5-4-5	10.5	—	—	—	—
	Pit 5-4-6	16.5	—	—	—	—
	Pit 5-4-7	14.1	—	—	—	—
<b>Pit 5-TW1</b>						
	—	—	Pit 5-TW1-M	10.2	Pit 5-TW1-L1	12.2
	—	—	Pit 5-TW1-MB	8.2	Pit 5-TWI-L2	12.3
	—	—	Pit 5-TW1-MB	2.9	—	—

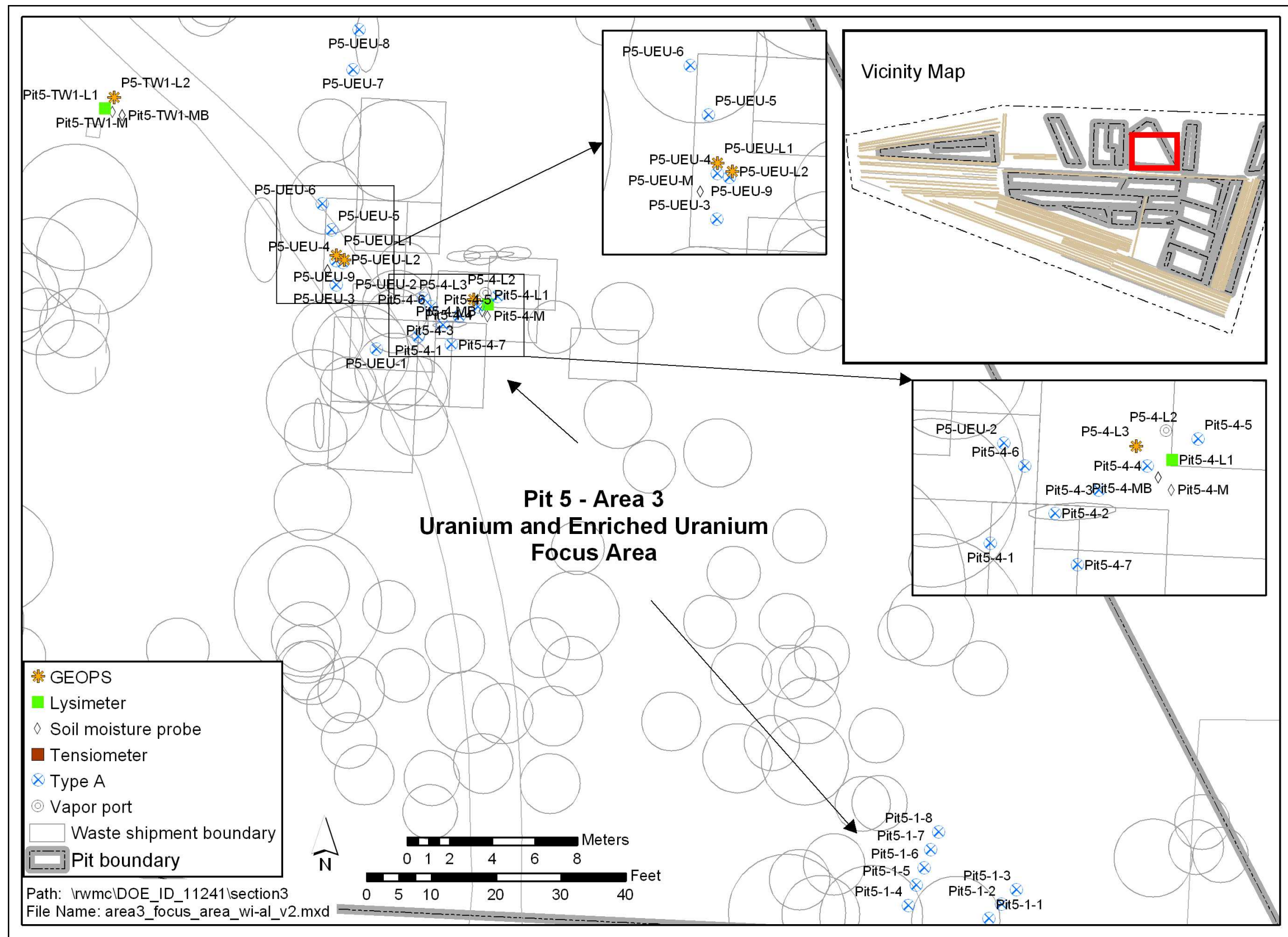


Figure 3-40. Probes installed in the Uranium and Enriched Uranium Focus Area in Pit 5.



**3.6.8.1 Probe Cluster Pit 5-1.** Type A probes were placed in an area shown by WasteOScope to contain two collocated disposals of U-233 waste from Rocky Flats Plant Building 881. Building 881 focused on enriched uranium manufacturing and recovery through the mid-1960s. However, Building 881 also housed numerous special projects, some of which involved U-233. Thirty-nine of the 370 drums reported for two disposals in the probing area contained U-233 waste from Building 881. Another three drums from these two disposals were reported in WasteOScope to contain U-233 from Building 771. Another important consideration in selecting this site for investigation is its disposal location along the southern perimeter of Pit 5. Confidence is higher for information on disposals near the pit boundary compared to information on disposals near the center of a large pit like Pit 5. Eight Type A probes were installed along two transects in Pit 5 (see Figure 3-40).

**3.6.8.2 Probe Cluster Pit 5-TW1.** Type B probes, including a lysimeter, were located in Pit 5 next to Lysimeter TW-1, where both U-236 and enriched uranium (i.e., anthropogenic uranium) previously had been detected in soil moisture collected from a depth of approximately 31 m (102 ft) (Roback et al. 2000). The area surrounding Lysimeter TW-1 also exhibits a topographic depression on the upper basalt surface (see Figure 3-40). This geologic feature may cause local infiltration around Pit 5 and Pad A to move in the direction of Lysimeter TW-1. Therefore, Type B probes were located in this area. The sampling port on the Type B Lysimeter Pit 5-TW1-L1 was completed at 3.7 m (12.2 ft) and is assumed to be where underburden and basalt contact. To date, however, Lysimeter Pit 5-TW1-L1 has not yielded soil-moisture samples, nor has the new GEOPS lysimeter (i.e., Pit 5-TW1-L2), which is located immediately north of Lysimeter TW1 at a depth of 3.7 m (12.3 ft).

**3.6.8.3 Probe Cluster Pit 5-4.** Seven Type A probes, two soil-moisture probes, and two lysimeters were installed in this area. Type A probes were placed in an area shown by WasteOScope to contain 14 of 147 drums that originated from Rocky Flats Plant Building 886, a building established for criticality testing on highly enriched uranyl nitrate. In addition, all but 16 of the drums in this disposal originated from Rocky Flats Plant uranium processing facilities (i.e., Buildings 881, 883, or 886), thus increasing the likelihood of detecting targeted uranium. WasteOScope was queried based on anecdotal information that two enriched-uranium-contaminated glove boxes and associated piping from Building 886 were buried in the SDA. A Rocky Flats Plant disposal was identified that described glove box deactivation, decontamination, and decommissioning-type waste and combustible waste originating from Building 886. This waste would be likely to contain significant concentrations of enriched uranium. Numerous spills containing highly enriched uranyl nitrate occurred in Building 886 and often were mopped up. If these mops (i.e., classified as combustible waste) were disposed of in the SDA, they would contain significant U-235 activity as well.

### **3.6.9 Activated Metal Investigations**

Activated metal investigations primarily focused on evaluating C-14. Carbon-14, as an activation product, is a by-product of reactor operations. Most C-14 inventory in the SDA is in activated metal. Some of this disposal inventory is in the form of reactor core components, including beryllium reflector blocks and end pieces from reactor cores. The remaining activity is mostly in ion-exchange resins. Typical C-14-bearing waste was buried in SVRs (or possibly in trenches) in the earlier years of operation (see Sections 3.1.2.4.2 and 3.2.1).

Type B probes were installed near soil vaults indicated by WasteOScope to contain C-14-bearing waste. These probes were placed to yield information about release and potential transport of C-14 in the subsurface. Carbon-14 can be transported in both vapor and dissolved phases. Moisture monitoring also was conducted near the vaults to assess the moisture state of surrounding soil. Two activated metal disposal sites were evaluated during the probing project investigation. One site, SVR 12, contained

activated stainless steel, and the other site, SVR 20, contained activated beryllium. Detailed criteria used to select each site for investigation are given in the Type B Probe Field Sampling Plan (Salomon 2001).

**3.6.9.1 Activated Metal (Stainless Steel) Investigation at Soil Vault Row 12.** Soil Vault Row 12 was selected to monitor activated stainless steel because disposal information indicated that the SVR had the target material and did not contain other interfering material (e.g., beryllium) that could greatly complicate subsequent analyses. WasteOScope data indicated disposal of highly irradiated waste at this site. Further investigation indicated that the waste contained disposals of highly irradiated stainless steel end pieces from spent fuel elements from Experimental Breeder Reactor II (Salomon 2001). This highly irradiated stainless steel probably was buried in scrap cask inserts that were open at the top and perforated on the bottom, allowing contact with surrounding soil (Salomon 2001). Because of shallow soil conditions at SVR 12, these disposals were made using an excavator in lieu of an auger rig to create the hole for burying the scrap cask inserts. The disposals of interest were handled remotely, using a free-air-transfer technique. As a result, exact positioning of the disposed waste was not possible.

Historical information in Salomon (2001) indicated that the SVR 12 disposal area was no deeper than 2 to 4 m (7 to 13 ft) below ground surface at time of disposal (Salomon 2001). However, because of subsequent flooding, RWMC Operations personnel placed approximately 3 m (9.8 ft) of fill in an area close to where these shipments were buried. Results of the probing did not support this because the greatest depth at which a probe could be installed at SVR 12 was only 3.8 m (12.5 ft) below ground surface (i.e., Probe SVR12-2-VP1). Specifics about waste disposal at SVR 12 and rationale used for selecting probe placement are given in Salomon (2001). Probe type and completion intervals for probes placed at SVR 12 are presented in Table 3-34. Physical placement of probes at SVR 12 is illustrated in Figure 3-41. Samples collected from vapor ports at SVR 12 are being analyzed for C-14, while samples collected from lysimeters are being analyzed for gamma spectroscopy and a suite of radionuclides (i.e., C-14, tritium, Nb-94, Ni-59, Ni-63, and Tc-99) to support modeling of activated-metal-release characterization.

**3.6.9.2 Activated Metal (Beryllium) Investigation at Soil Vault Row 20.** Six neutron-activated beryllium reflector blocks from the Advanced Test Reactor were buried in SVR 20 in 1993. The blocks contained significant tritium and C-14 activity. Section 3.9 summarizes beryllium disposals at SVR 20 and previous monitoring activities conducted nearby.

Before the SDA Probing Project, a monitoring array was established to characterize tritium migration and C-14 at SVR 20. The monitoring array installed as part of the SDA Probing Project augments monitoring that began in 1994. Vapor ports have been installed to enhance lateral monitoring for tritium and C-14 at a greater distance from the source than previously installed vapor ports. Moisture monitoring also is being conducted near the vault because the moisture state of the surrounding soil affects sampling and evaluation of soil-gas data. Probes installed for the SDA Probing Project investigation at or adjacent to SVR 20 are included in Table 3-34 and illustrated in Figure 3-41.

Vapor ports installed around SVR 20 are being sampled with a dedicated system for tritium sample collection. The system consists of a vacuum pump, control unit, and desiccant-filled moisture traps to monitor tritium in the vapor phase; this was not an objective of sampling at any other site.

Table 3-34. Clusters of probes supporting activated metal assessment.

Cluster Name	Probe Type							
	Tensiometer		Soil-Moisture Probe		Lysimeter		Vapor Port	
	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)	Probe Name	Port Depth (ft)	Probe Name	Port Depth (ft)
<b>SVR-12</b>								
	—	—	SVR-12-M	11.4	—	—	—	—
	—	—	SVR-12-MB	8.4	—	—	—	—
	—	—	SVR-12-MB	4.3	—	—	—	—
<b>SVR12-1</b>								
	SVR12-1-T3	10.8	—	—	SVR12-1-L1	11.1	SVR12-1-VP1	11.7
	SVR12-1-T2	8.4	—	—	SVR12-1-L2	5.8	SVR12-1-VP2	7.6
	SVR12-1-T1	3.6	—	—	—	—	SVR12-1-VP3	2.7
<b>SVR12-2</b>								
	—	—	—	—	—	—	SVR12-2-VP1	11.9
	—	—	—	—	—	—	SVR12-2-VP2	7.7
	—	—	—	—	—	—	SVR12-2-VP3	2.6
<b>SVR12-3</b>								
	—	—	—	—	—	—	SVR12-3-VP1	11.8
	—	—	—	—	—	—	SVR12-3-VP2	7.6
	—	—	—	—	—	—	SVR12-3-VP3	2.5
<b>SVR-20</b>								
	—	—	SVR-20-M	17.4	—	—	—	—
	—	—	SVR-20-MB	13.8	—	—	—	—
	—	—	SVR-20-MB	4.4	—	—	—	—
<b>SVR20-1</b>								
	SVR20-1-T3	16.4	—	—	—	—	—	—
	SVR20-1-T2	12.7	—	—	—	—	—	—
	SVR20-1-T1	8.3	—	—	—	—	—	—
<b>SVR20-3</b>								
	—	—	—	—	—	—	SVR20-3-VP1	6.3
	—	—	—	—	—	—	SVR20-3-VP2	12.9
	—	—	—	—	—	—	SVR20-3-VP3	15.0
<b>SVR20-5</b>								
	—	—	—	—	—	—	SVR20-5-VP3	17.2

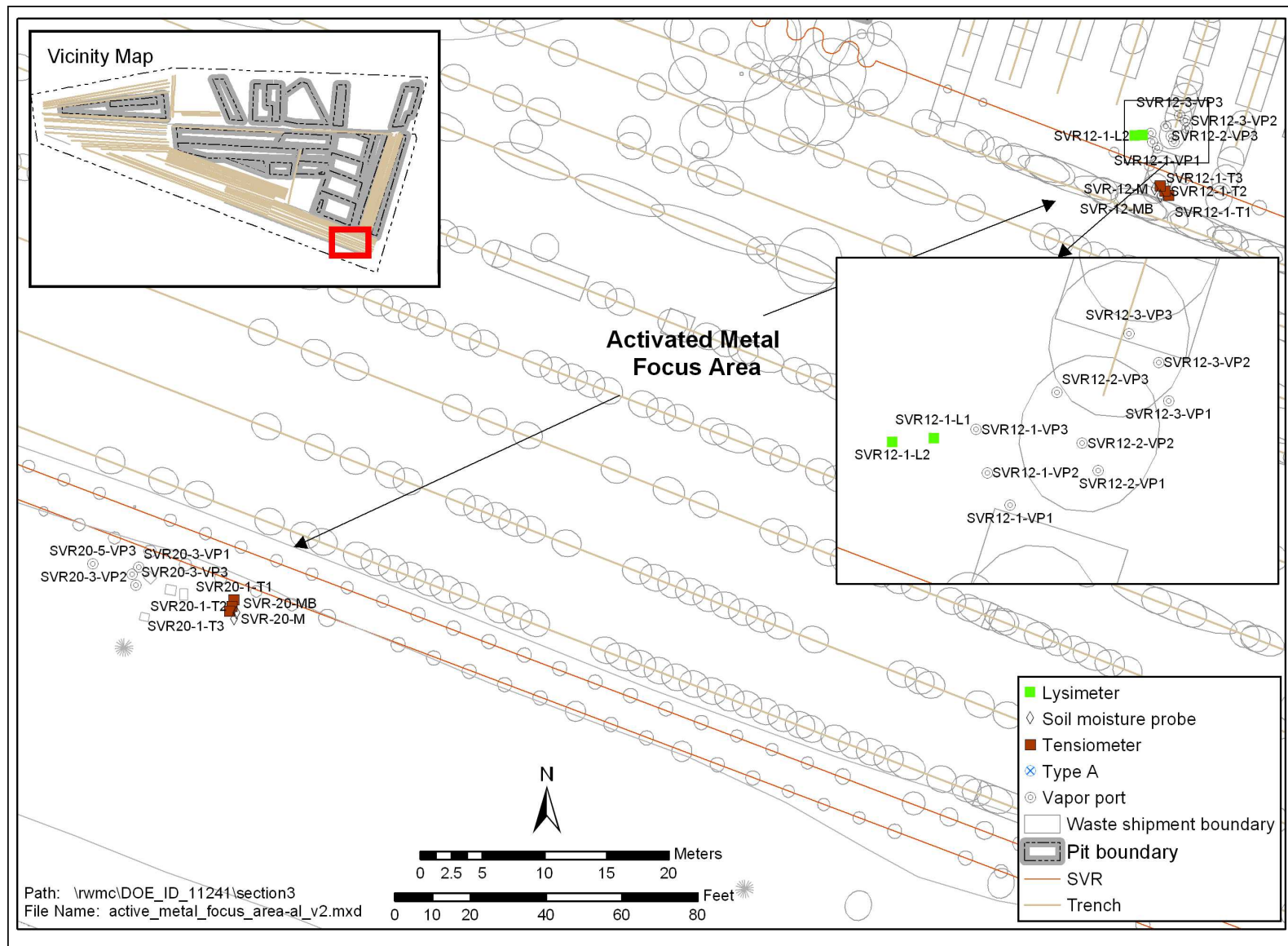


Figure 3-41. Probes installed in the Activated Metal Focus Area.



### 3.6.10 Waste Zone Moisture Monitoring Array

Three moisture monitoring transects and an additional array were established by pairing tensiometers and soil-moisture probes at various locations and depths in and immediately adjacent to some SDA pits. Primary purposes for these transects were (1) to identify the amount of water that infiltrates through the waste, (2) to investigate the effects of cover material on reducing infiltration, (3) to evaluate the influence of existing ditches on moisture movement, and (4) to determine whether local conditions could enhance contaminant release. Results of modeling are extremely sensitive to infiltration rates used in simulations. Not only does infiltration enable contaminant migration, it also influences the corrosion rate of metal containers and eventual release from the waste form. Though INL Site-specific infiltration rates had been implemented in the ABRA (Holdren et al. 2002) (see Section 5), the moisture monitoring transects were deployed to further validate the Operable Unit 7-13/14 comprehensive RI/FS model parameters and to enhance the infiltration data set for any future analysis of the SDA.

Three north-to-south-trending probe transects were installed in and adjacent to the northern side of Pit 4, and an additional array of probes was placed in the western end of Pit 10. In each Pit 4 transect, three clusters of probes were installed. The first cluster in each transect was installed near the drainage ditch north of Pit 4, the middle clusters were established near the northern boundary of the pit, and the third (southernmost) clusters were installed well into waste in Pit 4 (see Figure 3-42). In most probe clusters, three pairs of tensiometers and soil-moisture probe instruments were installed to monitor three different depths. As field conditions allowed, a tensiometer and soil-moisture detector were paired near the contact between the overburden and waste, near the middle of the waste zone, and near the contact between the waste and the underburden or near the basalt contact. Instruments in the center of the waste were installed with the tensiometer completed in the upper third of the waste zone and the soil-moisture detector completed lower in the middle of the waste zone to increase coverage in the waste. Final instrument completions in the moisture monitoring transects are given in Table 3-35.

The location of the transect known as MM1 was selected to monitor the effect of water that flows through a culvert under the east-west road (see Figure 3-42). The MM2 transect is centrally located along the northern edge of Pit 4. The MM3 transect is located just east of the I-3 monitoring well pair, which showed wet conditions above the B-C interbed at a depth of approximately 27 m (89 ft) during previous investigations.

An additional array of probes, MM4, was installed to form an array around the Depleted Uranium Focus Area in the western end of Pit 10. Some probes were located in a potential topographic depression on the underlying basalt surface (Salomon 2001). Several probes in this location are near a drainage ditch that borders the southwestern corner of Pit 10. The MM4-3 cluster is located in a slight topographic depression, while the MM4-2 cluster is located in an area that has higher relative surface elevation with good surface water run-off. This location is biased toward an area of suspected low infiltration to monitor moisture behavior in an area with less favorable infiltration potential.

Additional information was considered concerning upper basalt topography and locations where water tends to accumulate on the surface from snowmelt and precipitation (e.g., ditches) when selecting moisture monitoring locations. Upper basalt surface topography indicates a possibility for infiltrating water to move laterally toward Pits 4 and 10 from both north and south. Areas in the SDA that had significant ponding during a February 1995 thaw are shown in Figure 3-43 (Bishop 1996). This melting and water-accumulation pattern was similar to ponding that occurred in 1993, 1994, and 1996. Water temporarily standing in ditches nearly surrounding the perimeter of Pits 4, 6, and 10 is shown in Figure 3-43. Moisture monitoring probes were placed to determine the extent of lateral movement away from these ditches into the waste zone.



### 3.6.11 Type A Logging Results Used to Estimate Cover and Waste Zone Thickness

Results generated from nuclear logging of Type A probes have provided additional information to assess thicknesses of the soil cover and waste zone. Vertical waste zone thickness and boundaries were interpreted assuming that the waste zone contains less soil, more void spaces, and increased amounts of hydrogen, iron, and chlorine compared with overburden and underburden soil. Neutron-neutron moisture logging data were used to interpret the top and bottom of the waste zone in each Type A probe hole; this was based on the assumption that low density, void spaces, and the presence of neutron-absorbing elements found in typical waste can combine to cause sharp reductions in measured instrument response. Sharp decreases in the moisture tool response were first observed during Pit 9 logging in 1999. Decreased tool response occurred consistently at about the expected depth of the top of the waste zone at 1.2 to 1.8 m (4 to 6 ft) and indicated very low soil-moisture levels of about 0 to 5% by volume below that depth. In some cases, moisture tool response was observed to increase sharply near the bottom of the probe hole, returning to typical soil-moisture values. For these reasons, neutron-neutron tool response appears to be a valuable aid in determining vertical limits of the waste zone.

Neutron-neutron moisture logging data were the primary data set used to interpret vertical waste zone boundaries. Silicon, calcium, potassium, and thorium levels, measured using other logging tools, also were considered in cases where moisture tool response was ambiguous. Logging methods used for various waste zone indicators are provided in Table 3-36. Logging data from each probe were examined for sharp decreases and increases in neutron-neutron tool response. Decreases in tool response indicate the top of the waste, and increases in tool response indicate the bottom of the waste. Depths were chosen at the approximate middle of the decrease or increase and were rounded to the nearest 0.15 m (0.5 ft). Interpreted boundaries then were compared against contamination indicators (e.g., gross gamma, gross neutrons, and chlorine) to ensure consistency and to recognize signal interference.

Preliminary evaluations of logging data to determine overburden and waste zone thickness are summarized in Table 3-37. The waste zone thickness estimated in Table 3-37 may be greater than the value indicated because no lower waste boundary was recognized during interpretation of some logging results. In a few cases, the lower boundary of the waste zone was assumed to lie below the maximum logged depth; therefore, results are defined as minimum waste zone thickness. In some logs, no indicators of waste or presence of a waste zone were indicated. Data from these probes were excluded from thickness calculations because the respective data were likely from probes installed at pit boundaries and are not representative.

Overburden thickness estimates listed in Table 3-37 were compared to previous evaluations that estimated cover or overburden thickness. Section 3.1.5 provides information on overburden thickness summarized from Neupauer (1995) and Barnes (1989). Section 3.1.6 provides geophysical interpretation on overburden thickness estimates at selected pits within the SDA. Cover thickness estimates based on interpretations of Type A logging results correlate well with previously developed estimates.

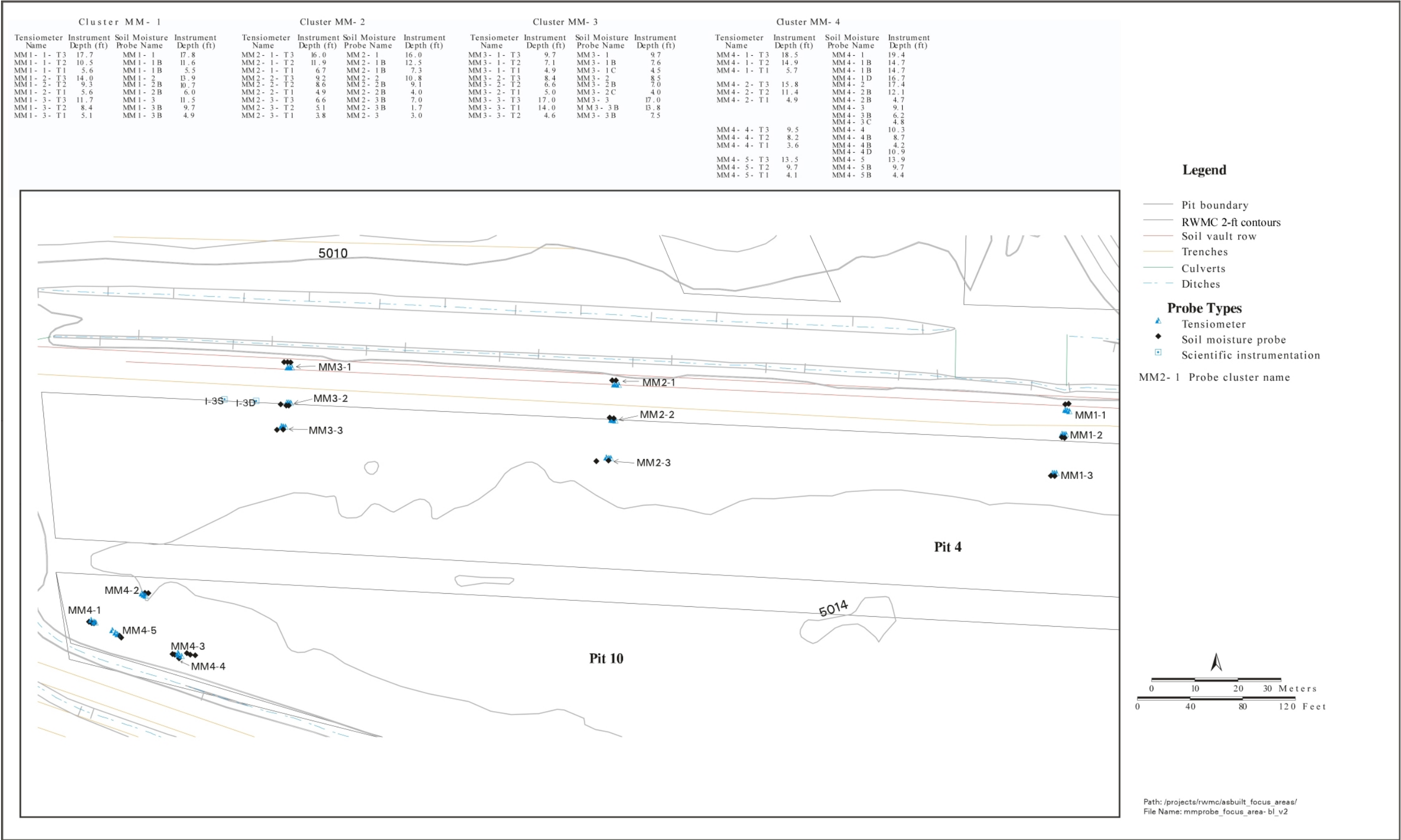


Figure 3-42. Probes installed to develop the waste zone moisture monitoring array.



Table 3-35. Clusters of probes in the waste zone moisture monitoring array at Pits 4 and 10.

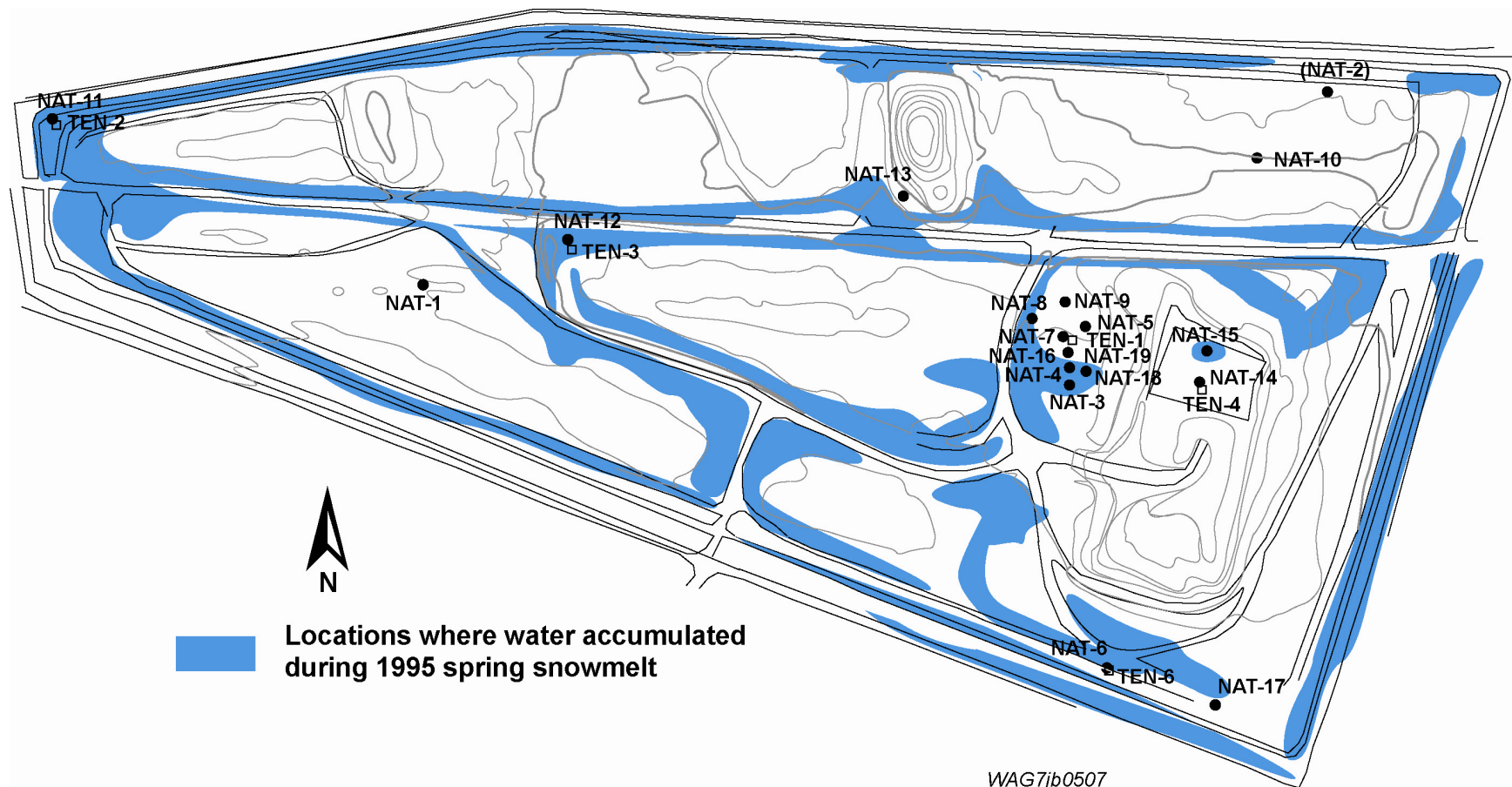
Cluster Name	Probe Type			
	Tensiometer		Soil-Moisture Probe	
	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)
<b>MM1-1</b>				
	MM1-1-T3	17.7	MM1-1	17.8
	MM1-1-T2	10.5	MM1-1B	11.6
	MM1-1-T1	5.6	MM1-1B	5.5
<b>MM1-2</b>				
	MM1-2-T3	14.0	MM1-2	13.9
	MM1-2-T2	9.3	MM1-2B	10.7
	MM1-2-T1	5.6	MM1-2B	6.0
<b>MM1-3</b>				
	MM1-3-T3	11.7	MM1-3	11.5
	MM1-3-T2	8.4	MM1-3B	9.7
	MM1-3-T1	5.1	MM1-3B	4.9
<b>MM2-1</b>				
	MM2-1-T3	16.0	MM2-1	16.0
	MM2-1-T2	11.9	MM2-1B	12.5
	MM2-1-T1	6.7	MM2-1B	7.3
<b>MM2-2</b>				
	MM2-2-T3	9.2	MM2-2	10.8
	MM2-2-T2	8.6	MM2-2B	9.1
	MM2-2-T1	4.9	MM2-2B	4.0

Cluster Name	Probe Type			
	Tensiometer		Soil-Moisture Probe	
	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)
<b>MM2-3</b>				
	MM2-3-T3	6.6	MM2-3B	7.0
	MM2-3-T2	5.1	MM2-3B	1.7
	MM2-3-T1	3.8	MM2-3	3.0
<b>MM3-1</b>				
	MM3-1-T3	9.7	MM3-1	9.7
	MM3-1-T2	7.1	MM3-1B	7.6
	MM3-1-T1	4.9	MM3-1C	4.5
<b>MM3-2</b>				
	MM3-2-T3	8.4	MM3-2	8.5
	MM3-2-T2	6.6	MM3-2B	7.0
	MM3-2-T1	5.0	MM3-2C	4.0
<b>MM3-3</b>				
	MM3-3-T3	17.0	MM3-3	17.0
	MM3-3-T1	14.0	MM3-3B	13.8
	MM3-3-T2	4.6	MM3-3B	7.5
<b>MM4-1</b>				
	MM4-1-T3	18.5	MM4-1	19.4
	MM4-1-T2	14.9	MM4-1B	14.7
	MM4-1-T1	5.7	MM4-1B	6.3
	—	—	MM4-1D	16.7

Table 3-35. (continued).

Cluster Name	Probe Type			
	Tensiometer		Soil-Moisture Probe	
	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)
<b>MM4-2</b>				
	MM4-2-T3	15.8	MM4-2	17.4
	MM4-2-T2	11.4	MM4-2B	12.1
	MM4-2-T1	4.9	MM4-2B	4.7
<b>MM4-3</b>				
	None installed	—	MM4-3	9.1
	—	—	MM4-3B	6.2
	—	—	MM4-3C	4.8

Cluster Name	Probe Type			
	Tensiometer		Soil-Moisture Probe	
	Probe Name	Instrument Depth (ft)	Probe Name	Instrument Depth (ft)
<b>MM4-4</b>				
	MM4-4-T3	9.5	MM4-4	10.3
	MM4-4-T2	8.2	MM4-4B	8.7
	MM4-4-T1	3.6	MM4-4B	4.2
<b>MM4-5</b>				
	—	—	MM4-4D	10.9
	MM4-5-T3	13.5	MM4-5	13.9
	MM4-5-T2	9.7	MM4-5B	9.7
	MM4-5-T1	4.1	MM4-5B	4.4



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Figure 3-43. Temporary accumulations of surface water at the Subsurface Disposal Area during February 1995.

Table 3-36. Logging methods used to interpret vertical waste boundaries.

Logging Method	Waste Zone Indication
Neutron-neutron (moisture)	Reduced response resulting from low density, void space, and neutron absorbing elements in waste
Passive spectral gamma	Reduced response resulting from reduced K-40 and Th-232 compared with native soil
Activated spectral gamma	Reduced response resulting from reduced silicon and calcium compared with native soil

Table 3-37. Estimates of overburden and waste zone thickness from Type A logging data.

Pit	Number of Probes Used to Determine Estimate	Overburden Thickness (ft)	Minimum Waste Zone Thickness (ft)
4	38	7.6	11.6
5	13	5.3	7.3
9	47	5.2	5.8
10	30	6.3	8.0

### 3.6.12 Continued Data Collection by Probes in the Subsurface Disposal Area

The last SDA GEOPS probes were installed in August 2004. Table 3-38 identifies those GEOPS lysimeters that have yielded sample volumes and provides information for samples collected to date.

Data generated by ongoing probe sampling and monitoring may be used for comparison to modeling results and, thus, assist in evaluating uncertainties associated with remedial decisions for the SDA. Data also could provide additional information useful during remedial design and remedial action. Data collection is planned to continue until 1 year after the Operable Unit 7-13/14 ROD is finalized (Holdren and Broomfield 2004).

Sampling using the GEOPS probe began in November 2004. Nine GEOPS instruments were sampled. Three were sampled as vapor ports; this required installing a higher vacuum on the instrument than was required for liquid. All three vapor ports produced several drops of liquid with the vapor. Four of the remaining six instruments produced liquid samples (i.e., one at 50 mL, two between 100 and 250 mL, and one at 260 mL). Sampling was suspended when the 260-mL sample was found to contain an unexpectedly high concentration of VOCs. Sampling resumed in March 2005, and a second round of sampling was conducted in June 2005.

The GEOPS lysimeters produced larger volumes of liquid than the Type B lysimeters, showing that the new sampling instrument works well and is collecting moisture from the waste zone (Meyer et al. 2005). Effectiveness of the new probe system was demonstrated during the first round of samples: 68% of GEOPS probes sampled produced liquid, and four of the six GEOPS lysimeter probes that were collocated with Type B lysimeters provided substantial liquid samples.



Table 3-38. Geologic and Environmental Probe System lysimeter information and sample volumes.

Sample Port	Depth		December 2004		March 2005		June 2005	
	(ft)	(m)	Approximate Volume (mL)	Color	Approximate Volume (mL)	Color	Approximate Volume (mL)	Color
DU-10-L3	7.9	2.41	0	—	—	—	Trace	—
DU-11-L1	17.1	5.21	0	—	—	—	Trace	—
DU-11-L2	11.3	3.45	180	Clear	—	—	20	Clear
DU-14-L3	13.3	4.05	260	Dark org-brn	—	—	150	Yellow-brown
DU-14-L4	13.2	4.02	—	—	0	—	—	—
DU-15-L1	16.1	4.91	120	Clear	—	—	10	—
DU-15-L2	16.1	4.91	40	Light brown	—	—	Trace	—
741-08-L3	15.2	4.63	—	—	0	NA	—	—
741-08-L4	7.7	2.35	—	—	0	NA	—	—
741-08-L5	15.1	4.60	—	—	0	NA	—	—
741-08-L6	11.0	3.35	—	—	460	Med brown	—	—
T3-EU-L1	14.1	4.30	—	—	3	Clear	10	—
T3-EU-L2	9.7	2.96	—	—	550	Clear	500	Clear
T3-EU-L3	23.1	7.04	—	—	20	Clear	5	—
P2-PU-L1	10.9	3.32	—	—	0	NA	0	—
P2-PU-L2	15.0	4.57	—	—	20	Light orange	Trace	—
P2-PU-L3	13.6	4.15	—	—	8	NA	0	—
P2-PU-L4	13.6	4.15	—	—	180	Light yel-org	0	—
HAL-2-L1	22.2	6.77	—	—	1	Clear	0	—
HAL-2-L2	22.0	6.71	—	—	2	Clear	0	—
T47-IF-L1	11.2	3.41	—	—	0	NA	0	—
T47-IF-L2	11.3	3.45	—	—	160	Light brown	20	Amber

Table 3-38. (continued).

Sample Port	Depth		December 2004		March 2005		June 2005	
	(ft)	(m)	Approximate Volume (mL)	Color	Approximate Volume (mL)	Color	Approximate Volume (mL)	Color
T48N-IF-L1	10.1	3.08	—	—	1	Clear	0	—
T48S-IF-L1	14.1	4.30	—	—	1	Clear	0	—
T50-IF-L1	12.7	3.87	—	—	2	Clear	20	Clear
P5 - 4 - L3	13.1	3.99	—	—	10	Clear	0	—
P5 - TW1 - L2	12.3	3.75	—	—	0	NA	0	—
P5 - UEU - L1	16.6	5.06	—	—	50	Light yel-brn	150	Clear
P5 - UEU - L2	13.4	4.09	—	—	25	Light yellow	Trace	
P6 - PU - L1	15.2	4.63	—	—	645	—	—	—
P6 - PU - L2	20.5	6.25	—	—	0	NA	—	—

The GEOPS probes have been in position for less than 1 year; to date, no limitations have been reported or observed on the sampled probes. Lysimeter samples appear to be of high quality and adequate volume to perform priority analyses. As with all lysimeters, results from organics and C-14 are potentially biased low because applying vacuum for a week and not collecting the sample immediately could cause volatilization and degradation of the sample.

### **3.6.13 Data Integration**

The Operable Unit 7-13/14 Probing Project spanned 5 years, from 1999 through 2004. The probing project generated enormous amounts of data, which have been used to assess disposal locations, subsurface conditions, contaminant distribution, and waste types. Detailed results from the Operable Unit 7-13/14 Probing Project are described by Josten and Okeson (2000); Holdren et al. (2002); INEEL (2002); Josten (2002); Myers et al. (2003, 2004); Josten (2005a); and Hubbell et al. (2005). Data obtained from the probing project were primarily qualitative; the benefit and uses of these data are summarized in the following subsections.

**3.6.13.1 Uses of Data from Type A Probes.** One hundred and ninety-one Type A probes were installed and logged in the SDA from December 1999 through August 2004. Locations for Type A probes were selected by first choosing a target waste stream, then evaluating inventory records for likely source areas, and finally reviewing surface geophysical data to refine the location (Salomon 2004). After installation, the location of each was surveyed, then a suite of logging tools (see Section 3.6.2) was lowered into the probe to collect data on the physical and contaminant characteristics within its sensing range. Type A logging data then were analyzed in terms of subsurface conditions, buried waste, and contaminant distribution.

Type A probes were useful in measuring thicknesses of the overburden, waste zone, and underburden. In general, depths were as expected, based on historical information (i.e., previous investigations, geophysical data, and RWMC maintenance records), but data from Type A probes were more precise:

- The neutron-neutron moisture-logging tool mapped vertical changes in the moisture content that correlated closely with each of these layers (Holdren et al. 2002)
- Cover thickness data corroborated with known cover data for pits where probes were installed (Holdren et al. 2002; Josten 2002)
- Depth to bedrock was obtained directly from probe refusal and correlated very well with known bedrock topography information (INEEL 2002; Holdren et al. 2002)
- The ability to interpolate bedrock topography improved with 191 new data locations.

Type A probe data also revealed the heterogeneous nature of the waste zone (Josten 2002, 2005b; INEEL 2002). For Pits 4, 5, 9, and 10, Type A probe data confirmed inventory records and historical photographic information that waste was randomly buried and covered, and that voids, soil, and waste were randomly distributed. Probes placed several feet from each other routinely encountered different waste types and subsurface conditions. However, logging data from Type A probes in Pit 2 clearly indicated that waste was stacked in drums, with minimal soil around the waste (Josten 2005a).

Data from Type A probes also correlated very well with surface geophysical data and inventory records. Surface geophysical data were used to evaluate pit boundaries, locate metallic objects, and map suspected waste streams and waste types in pits and trenches. Many of these interpreted subsurface conditions were confirmed when compared with logging data from Type A probes (Josten 2002; INEEL 2002).

Type A nuclear logging data were used to define unique combinations of various radioisotopes, trends of radioisotopes, or unique characteristics used to target Type B probes. Each targeted waste stream (i.e., depleted uranium, enriched uranium, organic sludge waste, plutonium-contaminated waste, or americium and neptunium waste) had different radioisotopes or different ratios of radioisotopes that could be used to crudely identify, or fingerprint, the waste. For example, Waste Area Group 7 personnel were able to discriminate between graphite molds and filter waste during the Pit 9 Campaign 2 using fingerprint radioisotope data and surface geophysics (INEEL 2002).

Trends of the same radioisotope also were used to define location, relative concentration, and waste type. In some instances, the azimuthal logging tool (see Section 3.2.10.1) was used to define the direction of a target waste in relation to the Type A probe. Unique characteristics of the waste zone also were learned, including the following:

- Unusually large logging responses were attributed to voids in the subsurface
- Moisture log data showed low apparent values throughout areas having elevated chlorine values
- Silicon, calcium, and potassium were reliable indicators of soil
- Heterogeneity of radionuclide distribution were clearly identified with the azimuthal logging tool
- Vertical migration of radionuclides into the underburden is limited.

Using these interpretations, Type B probes then were installed both vertically and horizontally to optimize sensors and sampling ports to monitor specific waste types or within the soil overburden, layers of soil within the waste, or underburden.

**3.6.13.2 Uses of Data from Type B Probes.** Data from Type B probes were used to characterize the physical nature of the subsurface, moisture trends, and contaminant movement. Visual probes were very useful to understand waste layer moisture trends (Myers et al. 2003, 2004; Josten 2002). Video logs showed the appearance of different waste types. When combined with logging data of adjacent Type A probes, video probes provided a better understanding of data from Type A probes. For example, physical features (e.g., a void, soil layers, unique waste, or presence of moisture) could be seen in the visual probe, and correlation could be seen in the nuclear logging data.

Vapor probe data were useful in characterizing organic sludge and in understanding VOC behavior within the waste. These data also were coupled with Type A probe data to understand better VOC distribution and behavior (Miller and Varvel 2005; Myers et al. 2003, 2004).

Little opportunity was available to integrate Type B moisture probe data because the soil-moisture, lysimeter, and tensiometer probes did not provide much useable data for 2002 through 2004 (Myers et al. 2003, 2004; Hubbell et al. 2005). Reasons for unusable data from these instruments include:

- Lack of precipitation
- Heterogeneous waste

- Sonic drilling technique (i.e., vibration and stress) causing failure of sensors and probes
- Instrument design.

The Type B lysimeter probe was redesigned as the GEOPS in 2004. The new probe was much more successful than its predecessor in collecting water samples in November 2004 after a wet fall. A full assessment of the new Type B lysimeter probe cannot be offered because its use was restricted after water samples exceeded safe handling levels for radiation.

**3.6.13.3 Summary of Integrated Data.** Data from Type A and B (i.e., GEOPS) probes, in combination with other investigations, were useful in providing characterization information about specific focus areas of interest in the SDA. These focus areas included the Organic Sludge Focus Area, Americium/Neptunium Focus Area, and Depleted Uranium Focus Area, which are further discussed in this section. Additionally, a discussion of the detailed investigation of the Pu-239 contamination surrounding the Location P9-20 probe is presented. Other focus areas included the Enriched Uranium Focus Area, Plutonium Focus Area, and Uranium and Enriched Uranium Focus Area.

**3.6.13.3.1 Organic Sludge Focus Area**—Data from Type A and B probes, in combination with other investigations, were useful in characterizing the Organic Sludge Focus Area located in the eastern end of Pit 4. Surface geophysical data defined areas of low metallic waste, high metallic waste, and no metallic waste, which correlated with SDA inventory records for organic sludge drum waste, other types of drummed waste, and debris, respectively (INEEL 2002). Subsequent Type A probes confirmed boundaries of organic waste relative to adjacent debris and other drummed waste (INEEL 2002). The n-gamma logging data for chlorine from Type A probes also correlated well with plume boundaries of historical soil-vapor data. Several mapped areas of elevated VOCs were shown to directly correlate to either large concentrations of buried organic sludge, areas of thinner overburden, or shallow waste suspected in the cover material (INEEL 2002) (see Figure 3-44). Figure 3-45 provides locations of the probing project focus areas. Data from Type B vapor probes installed within the Organic Sludge Focus Area also validated assumptions about historical disposal information from the 1960s (Myers et al. 2003; Miller and Varvel 2005).

**3.6.13.3.2 Americium and Neptunium Focus Area**—The Americium and Neptunium Focus Area is located near the center of Pit 10. This area was targeted based on historical disposal records indicating that a high percentage of Series 741 sludge from Rocky Flats Plant was buried in this location (Becker et al. 2000). Detailed analyses of data from original Type A probes in this focus area indicate an abundance of Am-241 and Np-237 that could not be attributed simply to decay of weapons-grade plutonium. These relatively high activities of Am-241 and Np-237 measured in the 741-numbered probes are concluded to have come from separate sources of Am-241 and Np-237 (Mandler and Giles 2000). Probe 741-08 was identified for further investigation, including placing a cluster of Type B lysimeters and Type A probes around Location 741-08 (see Figure 3-45). Americium-241, Np-237, and Pu-239 were detected in Probe 741-08 from the 2 to 4.8-m (7 to 16-ft) depth interval; relative activities peaked at a depth of 2.4 (8 ft). Data from the other Location 741-08 Type A probes (i.e., Probes 741-08-A and 741-08-B) did not show positive detections for Am-241 or Np-237, indicating that targeted shipments were not within the effective measuring range (i.e., 0.3 to 0.48 m [12 to 18 in.]) of the probes. Of the six lysimeters placed near Location 741-08, two have yielded liquid samples: Lysimeters 741-08-L1 and 741-08-L6. Results from Lysimeter 741-08-L1 (i.e., 4.65 m [15.24 ft] deep) are consistent with data from the Type A probe; Am-241 and Np-237 activity are considerably higher than expected from decay of weapons-grade plutonium, confirming that the waste contains separate sources of Am-241 and Np-237 (e.g., Series 741 sludge).

Chlorine slice and plume with moisture  
represented by probe bores - 743 Focus Area

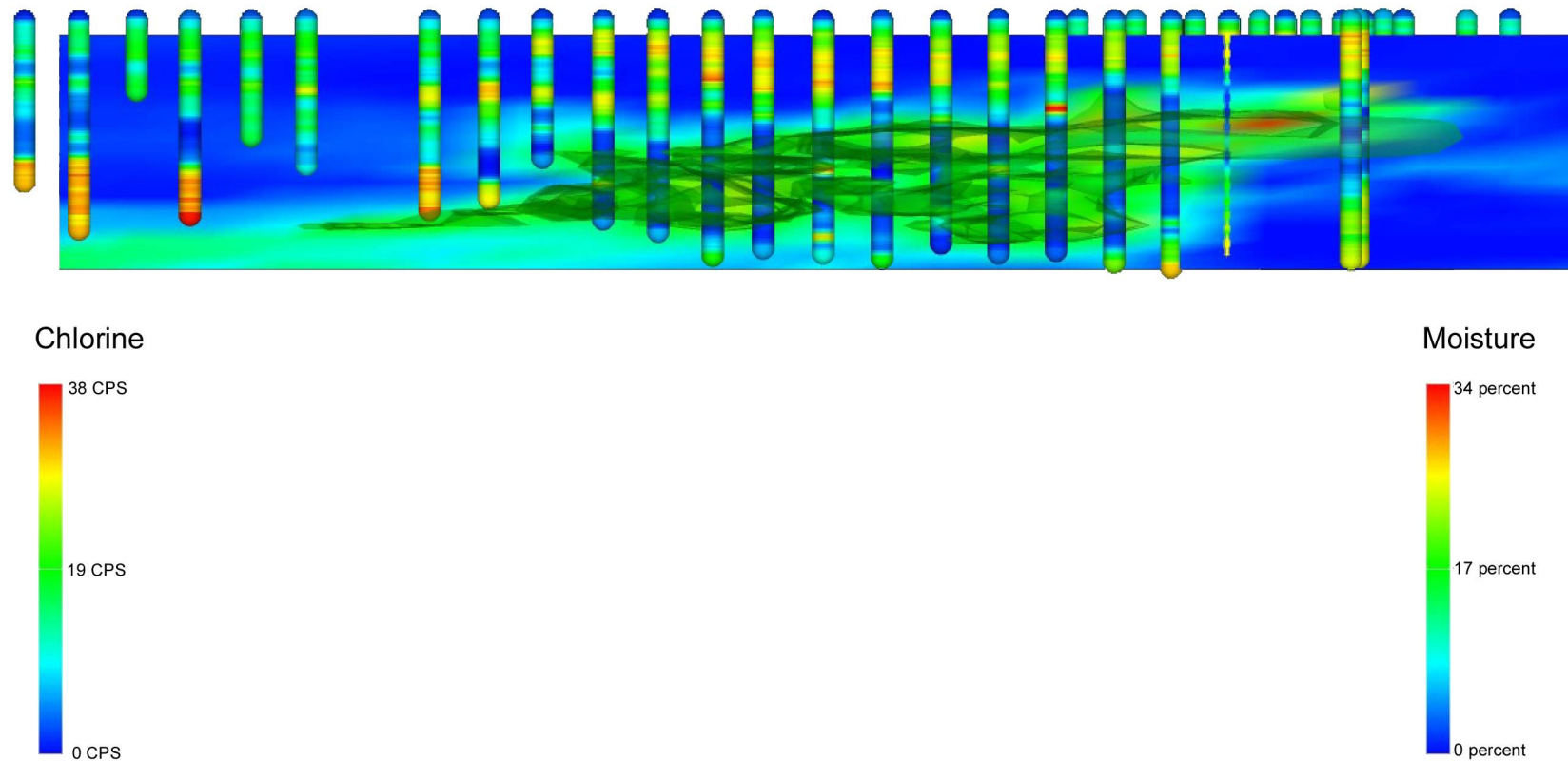


Figure 3-44. Chlorine slice and plume with moisture represented by probe bores in the Series 743 Focus Area.

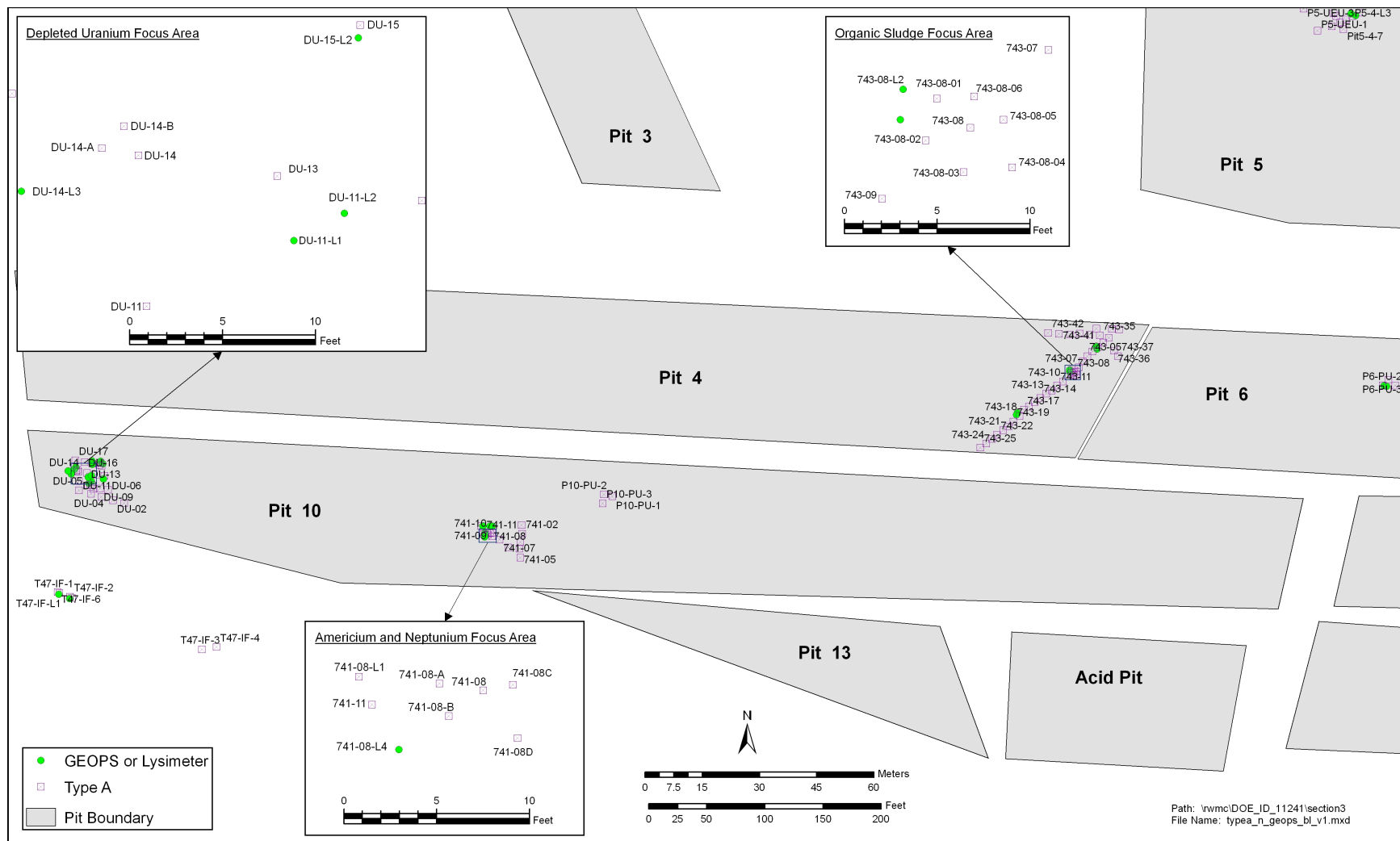


Figure 3-45. Probing project focus areas at the Subsurface Disposal Area.



**3.6.13.3.3 Depleted Uranium Focus Area**—The Depleted Uranium Focus Area is at the far western end of Pit 10. This area was targeted, based on historical records indicating that (1) depleted uranium was the primary waste stream buried in this part of the pit and (2) enriched uranium also was buried in this area. However, collocation of enriched uranium and depleted uranium waste streams could prevent discriminating between these specific waste types by Type A probe nuclear logging tools.

The purpose of selecting locations in the Depleted Uranium Focus Area for initial Type A probes was to resolve the discrepancy between location of waste as identified by historical disposal records and location shown by surface geophysics. Eight Type A probes were placed in two transects in an attempt to locate the buried depleted uranium waste (Becker et al. 2000). Analysis of logging data showed sparse distribution of depleted uranium across the two transects. Based on these results, an additional nine Type A probes were placed to define more closely the extent of the depleted uranium waste disposal area. Of the second set of Type A probes, Location DU-14 yielded the highest apparent concentration of U-238 with a value of 17,300 pCi/g, almost 10 times the previous high concentration in Location DU-03 (see Figure 3-45). A widespread distribution of uranium through a depth interval ranging from 1.8 to 4.6 m (6 to 15 ft) also was noted in Location DU-14. Based on these observations, two additional Type A probes and four lysimeters were placed near Location DU-14 at a depth of approximately 4 m (13 ft), which is near the bottom of the waste layer. Liquid samples collected from Lysimeter DU-14-L3 have not yielded high concentrations of radionuclides, and uranium has been below the method detection limit; however, high concentrations of VOCs have been observed. Logging results from Probes DU-14-A and DU-14-B show that uranium distribution is similar to that found in Location DU-14; however, apparent concentrations are approximately 75% less. Locations DU-11 and DU-15 were also identified for placement of Type B or GEOPS lysimeters. Lysimeters near these probes were placed at depths of 3.4 and 4.9 m (11.3 and 16.1 ft), respectively, which are just below the waste zone interval containing U-238 as indicated by the Type A logs of the corresponding probes.

**3.6.13.4 Summary of Installed Data for Location P9-20 Investigation.** Data from Type A and B probes were used to characterize elevated levels of plutonium reported at Location P9-20 in Pit 9. During the Pit 9 12 × 12-m (40 × 40-ft) campaign, 20 Type A probes were installed in a grid pattern over an area suspected of containing plutonium waste from Rocky Flats Plant (Josten and Okeson 2000). One Type A probe encountered elevated levels of Pu-239, with values that were orders of magnitude greater than any other probe. Eight more Type A probes were installed just north of Location P9-20 during Pit 9 Campaign 1 in an effort to bound the area of elevated plutonium (Josten and Okeson 2000). Logging data showed that the elevated source of Pu-239 was only at Location P9-20, and the waste type was most likely plutonium-contaminated graphite waste at 2 m (6 ft) and Series 741 waste at 2.7 to 3 m (9 to 9.8 ft). Azimuthal logging data confirmed that two sources were close to Location P9-20. Six additional Type A probes were installed in a circular, 1-m (3-ft) -radius pattern around Location P9-20 and logged. These data were then modeled to define the volume and concentration of the Pu-239 source(s) present. A Type B visual probe was installed within 30 cm (12 in.) of Location P9-20 and showed that (1) the Type A probe at Location P9-20 had penetrated a drum, (2) large voids were adjacent to the elevated Pu-239 areas, and (3) soil was abundant around the probe (Jewell, Reber, and Hertzog 2002). Further analysis to support Pit 9 excavation showed that distribution of waste types identified by Pit 9 Type A probes correlated very well with historical inventory records (Jamison and Preussner 2002). The excavation did encounter waste types and subsurface conditions predicted by data from the Type A probes and revealed that the probe at Location P9-20 had pierced a drum containing graphite waste as shown in Figure 3-13 (DOE-ID 2004a; Olson 2004). The Location P9-20 investigation demonstrated that quantitative analysis of radionuclide concentrations and distribution is possible but requires extensive collection and analysis of data to reduce uncertainty to acceptable levels. This level of analysis is not feasible on a large scale, as Miller and Varvel (2005) showed during VOC analysis.

### 3.7 Actinide Retardation Studies

A series of experiments was conducted in laboratories at Clemson University (Fjeld, Coates, and Elzerman 2000) to measure radionuclide retardation parameters for application to risk assessment at the SDA. The experiments focused on actinide elements (i.e., uranium, neptunium, plutonium, and americium). Experiments were conducted mainly with composite interbed sediment, with a few studies conducted with basalt. Recently, experiments were conducted on material collected from discrete intervals in sedimentary interbeds. Laboratory studies consisted of batch measurements, column elution experiments, and measurement of adsorption isotherms. Test solutions were made to resemble the chemistry of groundwater near the SDA.

The objective of these experiments was to quantify partitioning of radionuclides between vadose zone water and sedimentary interbed material from the SDA. Most experiments were designed to measure a linear, reversible partition coefficient referred to as  $K_d$ . The  $K_d$  parameter is defined as the ratio of the quantity of a contaminant sorbed onto a solid phase (e.g., sediment or basalt) to the quantity of the contaminant remaining in the water, as shown in Equation (3-1):

$$K_d = \frac{C_s}{C_w} \quad (3-1)$$

where

- $K_d$  = linear, reversible partition coefficient (mL/g)
- $C_s$  = quantity of the contaminant on a solid (pCi/g, mg/g)
- $C_w$  = quantity of the contaminant in water (pCi/mL, mg/mL).

In column experiments, retardation of the movement of a radionuclide relative to the rate of water movement through the column was measured. Retardation of a radionuclide is related to the  $K_d$  as shown in Equation (3-2):

$$R = 1 + \frac{\rho}{\theta} K_d \quad (3-2)$$

where

- $R$  = retardation factor (unitless)
- $\rho$  = bulk density of sediment in the column (g/mL)
- $\theta$  = water content of the column (mL/mL).

Rearranging Equation (3-2),  $K_d$  can be calculated using Equation (3-3):

$$K_d = \frac{\theta}{\rho} (1 - R) \quad (3-3)$$

Equation (3-3) is used in this section to calculate  $K_d$  values from retardation factors reported from column experiments.

Between 1993 and 1995, a series of batch adsorption experiments and a few column elution experiments were conducted at Clemson University and at the INL Site using uranium, plutonium, and americium (Newman et al. 1996). Nine sedimentary interbed samples from multiple depths in Wells M6S and M7S, which were drilled near RWMC, were mixed to form a composite interbed material. The composited material was sieved to a size less than 0.25 mm. A few tests were conducted on a crushed INL Site basalt and an intact core of basalt. Tests were conducted in a synthetic groundwater with a composition similar to groundwater in the Snake River Plain Aquifer near RWMC (pH = 8.0;  $\text{HCO}_3^- = 96.6 \text{ mg/L}$ ). Column tests were conducted in packed columns 20 to 25 cm (8 to 10 in.) long. Batch partition experiments were conducted following the “Standard Method for Distribution Ratios by the Short-Term Batch Method” (ASTM 2001).

For column tests on basalt, uranium transport was reasonably well explained by a  $K_d$  type model, with about 100% recovery. Retardation, as defined in Equation (3-2), was about 2 for uranium, translating to a  $K_d$  of about 0.2 to 0.3 mL/g (see Table 3-39). Plutonium (total) and americium showed an enhanced mobility fraction of about 2%, with a retardation factor, as defined in Equation (3-2), of about 2 for crushed basalt columns. The remaining 98% of plutonium and americium did not emerge from the crushed basalt column, indicating that retardation exceeded 200. Based on the bulk density and porosity of the column, a retardation value greater than 200 gives a  $K_d$  value greater than 60 mL/g (see Table 3-39). In columns containing packed composited interbed sediment, uranium was slightly retarded during transport through the column, with 90 to 100% recovery of the tracer. Retardation ranged from 30 to 43, yielding  $K_d$  values between 7 and 10 mL/g (see Table 3-39). Plutonium and americium showed a very small fraction of enhanced transport, with more than 99% of the plutonium and americium remaining in the column, indicating a  $K_d$  greater than 48 mL/g.

Table 3-39. Summary of batch and column measurements of americium, plutonium, and uranium partition coefficients.

Isotope	Media	Batch $K_d$ (mL/g)	Column $K_d$ (mL/g)	Enhanced Mobility $K_d$ (mL/g)
Americium	Basalt	70 to 280	>60 <sup>a</sup>	0.2
Americium	Interbed	450 to 1,100	>48 <sup>a</sup>	Present <sup>b</sup>
Plutonium(V)	Basalt	70 to 130	—	—
Plutonium(V)	Interbed	5,100 to 7,900	—	—
Plutonium(V)	Surface soil	7,800 to 22,000	—	—
Plutonium(VI)	Basalt	12 to 24	—	—
Plutonium(VI)	Interbed	110 to 690	—	—
Plutonium(VI)	Surface soil	1,800 to 4,900	—	—
Plutonium (total)	Basalt	—	>60 <sup>b</sup>	0.25
Plutonium (total)	Interbed	—	>48 <sup>b</sup>	Present <sup>b</sup>
Uranium	Basalt	4 to 6	0.2 to 0.3	Not present
Uranium	Interbed	3 to 6	7 to 10	Not present

Note: Information in this table is from Newman et al. (1996).

a. Breakthrough did not occur; therefore, it is known only that the  $K_d$  is greater than the reported value.

b. An enhanced mobility fraction was present, but at too small a concentration to quantify a  $K_d$ .

A wide range of  $K_d$  values was measured for plutonium in batch adsorption experiments (see Table 3-39). For comparable material, higher  $K_d$  values were measured when the plutonium initial oxidation state was plutonium(V) and lower values when the starting oxidation state of plutonium was plutonium(VI) (Newman et al. 1996) (see Figure 3-46). Most batch partition experiments were conducted at a water-to-solid weight ratio of 4:1 as specified in the “Standard Method for Distribution Ratios by the Short-Term Batch Method” (ASTM 2001). In the vadose zone, the water-to-solid weight ratio is closer to 1:5, as calculated with a soil bulk density of  $1.5 \text{ g/cm}^3$  and a moisture content of  $0.3 \text{ cm}^3/\text{cm}^3$  (Barraclough et al. 1976), which is substantially different from laboratory results. Oxidation-state determinations on plutonium in batch experiments showed that when the solid-to-liquid ratio in experiments more closely resembled natural conditions in the vadose zone, plutonium(V) and plutonium(VI) were reduced to plutonium(IV).

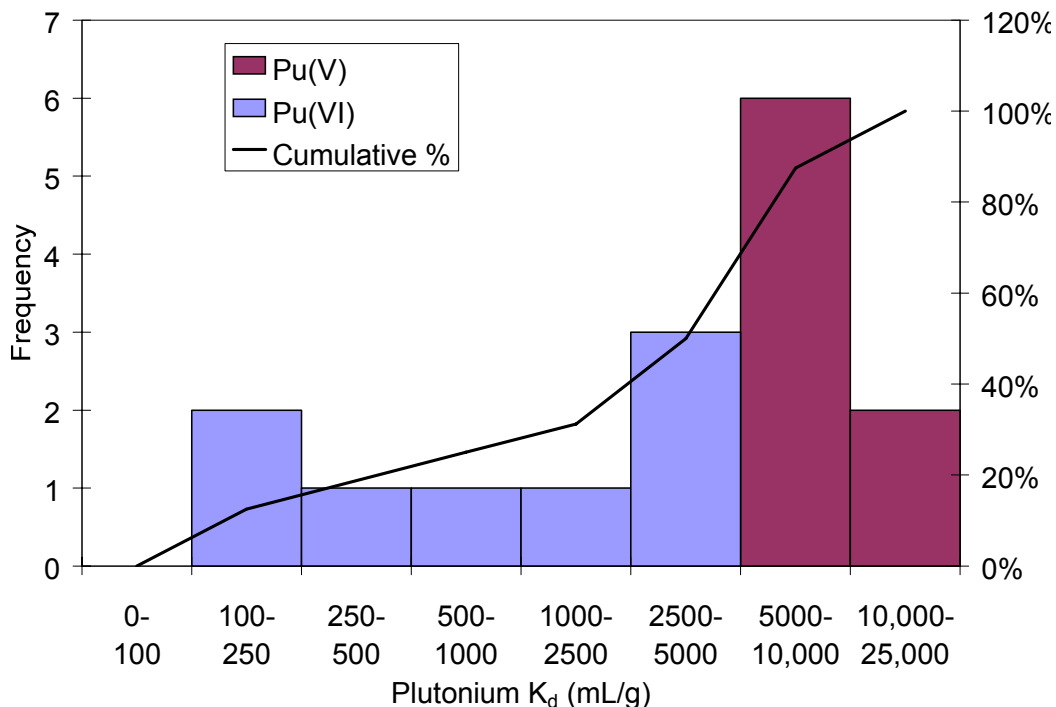


Figure 3-46. Frequency distribution of batch plutonium  $K_d$  values differentiated by the valence state of plutonium in the starting solution.

Additional column studies were conducted at Clemson University on thorium, uranium, neptunium, plutonium, and americium to further define transport mechanisms and parameters (Fjeld, Coates, and Elzerman 2000). These studies were conducted on composite interbed material in a synthetic water solution designed to resemble perched water at RWMC ( $\text{pH} = 8.2$ ;  $\text{HCO}_3^- = 750 \text{ mg/L}$ ). All of the experiments were column-elution experiments. This series of tests included variations in water chemistry to evaluate effects of actinides complexing with ligands in solution.

Trivalent and tetravalent actinides, americium(III), thorium(IV), and plutonium(IV) were found to have a very small (i.e., less than 0.01%) enhanced mobility fraction with a retardation factor between 1 and 6 and a  $K_d$  less than  $0.5 \text{ mL/g}$ . Americium(III) and plutonium(IV) showed a continuous small breakthrough during the experiment. However, 99.9% of the americium(III), 98.8% of the plutonium(IV) and 99.9% of the thorium(IV) were retained in columns with a  $K_d$  greater than  $250 \text{ mL/g}$  (see Table 3-40). Plutonium(V) showed the same behavior as plutonium(IV), with a small fraction being released from the column with little retardation, and 99.8% of plutonium(V) remaining in the column. Plutonium(VI) was not tested.

Table 3-40. Summary of retardation and  $K_d$  values measured in column experiments.

Element	Special Test Conditions	Column Retardation Factor	Column $K_d$ (mL/g)	Fraction of Initial Spike Retained in the Column	Enhanced Mobility (fraction released/pore volume)	Effective $K_d$ of Enhanced Mobility Fraction (mL/g)
Americium	—	>1,000	>250 <sup>a</sup>	0.999	7.9E-5	4.5
Americium	With EDTA <sup>b</sup>	16	4	0.50	7.7E-5	Continuous <sup>c</sup>
Thorium	—	>1,000	>250	0.999	2.6E-5	Continuous
Thorium	With EDTA	23	5.75	0.73	2.3E-5	Continuous
Plutonium(IV)	—	>1,000	>250	0.98	6.2E-6	Continuous
Plutonium(IV)	With EDTA	35 to 79	8.8 to 20	0.42	1.7E-4	Continuous
Plutonium(V)	—	>1,000	>250	0.999	1.7E-4	Continuous
Plutonium(V)	With EDTA	>1,000	>250	0.999	3.6E-5	Continuous
Plutonium(V)	Oxidized sediment <sup>d</sup>	>1,000	>250	0.999	Not determined	—
Neptunium	—	97 to 156	24 to 39	0.32 to 0.60	1.5E-4	0.3 to 0.9
Neptunium	With EDTA	58 to 80	14.5 to 20	0.33 to 0.43	1.5E-4	0.6
Neptunium	No carbonate <sup>e</sup>	195 to 310	49 to 78	0.19 to 0.33	Not present	NA
Neptunium	Oxidized sediment	265 to 434	66 to 108	~0	Not present	NA
Uranium	—	3.2 to 8.2	0.5 to 2	0.04 to 0.08	Not present	NA
Uranium	With EDTA	Not tested	—	—	—	—
Uranium	No carbonate	560 to 690	140 to 170	0.55	1.3E-4	3

a. Breakthrough did not occur, and the only conclusion that can be drawn is that the  $K_d$  is greater than the reported value.

b. Simulated perched water with EDTA added.

c. An enhanced mobility fraction was present, but release was more or less continuous, not as a discrete peak.

d. Hydrogen peroxide was used to oxidize the sediment column before the test.

e. Carbonate was left out of the recipe when the simulated perched water was mixed.

EDTA = ethylenediaminetetraacetic acid

Neptunium experiments on the column were conducted using neptunium in the neptunium(V) valence state. About half of the neptunium migrated through the columns with a retardation factor of 100 and a  $K_d$  of 25 mL/g (see Table 3-40). The other half of the neptunium did not emerge from the column. Fjeld, Coates, and Elzerman (2000) hypothesized that some of the neptunium was being reduced from the +V to the +IV valence state in the column. A column was oxidized with hydrogen peroxide, and a breakthrough test was repeated. All of the neptunium was recovered, indicating that reduction was a factor in neptunium transport. A similar experiment with plutonium(V) did not result in breakthrough of plutonium. Reduction of plutonium in the soil is strong enough that, even after treatment of the soil with hydrogen peroxide, the soil can reduce plutonium(V) to plutonium(IV).

Column experiments were conducted using the hexavalent form of uranium. Uranium(IV) can be expected to have similar geochemical characteristics as other quadravalent actinides (Seaborg and Loveland 1990). Under reducing conditions, where uranium(IV) predominates, uranium would be strongly bound to sediment with  $K_d$  values greater than 250 mL/g, similar to values measured for thorium and plutonium(IV). Hexavalent uranium was completely recovered from columns with a retardation factor of 3.2 to 8.2 and a  $K_d$  from 0.54 to 1.8 mL/g (see Table 3-40). Water chemistry composition experiments showed that complexing uranium with carbonate ion was a very important factor in uranium transport. When carbonate ions were removed from the synthetic groundwater, the retardation factor jumped to 560 to 690 and the  $K_d$  jumped to 140 to 170 mL/g.

Complexing with organic complexing agents also was found to appreciably enhance migration of actinides. Quadravalent actinides were not mobile in the absence of complexing agents. When EDTA was added to test solutions, 27 to 61% of the quadravalent actinide became mobile with retardation factors from 16 to 79 and  $K_d$  values from 4 to 20 mL/g (see Table 3-40). Organic complexing agents were buried in the SDA (INEEL 1998), including unknown quantities of EDTA contained in Rocky Flats Plant TRU waste. According to the Historical Data Task, a reasonable upper limit on the unknown quantity of EDTA from Rocky Flats Plant is  $7.1 \times 10^4$  kg (INEL 1995a, Table 4-1). Hypothetically, presence of these agents could enhance mobility of actinides. However, because detections of actinides in the vadose zone are relatively sparse, the hypothesis has not been confirmed by results of RWMC vadose zone monitoring (see Section 4).

During the fall of 2000, adsorption isotherms for uranium and neptunium were measured on 14 discrete interbed samples collected from boreholes drilled inside and adjacent to the SDA (Grossman et al. 2001). An adsorption isotherm is measured by a series of batch adsorption experiments carried out with different initial solution concentrations. If adsorption follows a linear adsorption isotherm ( $K_d$ ), the ratio of sorbed to dissolved concentrations will be the same at all concentrations. Sediment samples from both the B-C and C-D interbeds were assessed. Experiments were again conducted in synthetic groundwater (pH = 8.2;  $\text{HCO}_3^- = 220$  mg/L).

For neptunium and uranium, isotherms showed distinct nonlinearity. However, a good fit for both actinides was obtained using a Freundlich isotherm (see Figure 3-47). The Freundlich isotherm is defined by the following equation:

$$C_s = K_f C_w^n \quad (3-4)$$

where

- $K_f$  = Freundlich adsorption coefficient (mL/g)
- $n$  = Freundlich exponential coefficient (unitless).

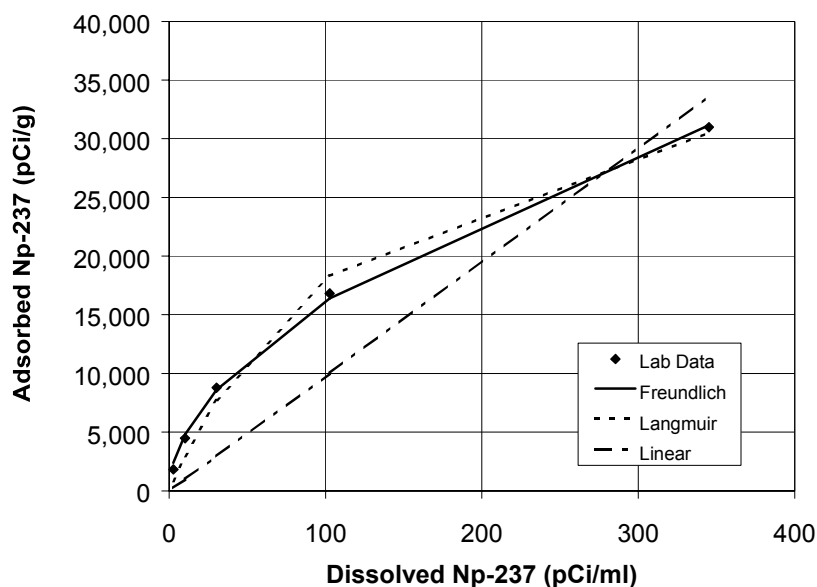


Figure 3-47. Neptunium adsorption on Sample I1S-INEEL-109 comparing the fit of Freundlich, Langmuir, and linear isotherms to the laboratory data.

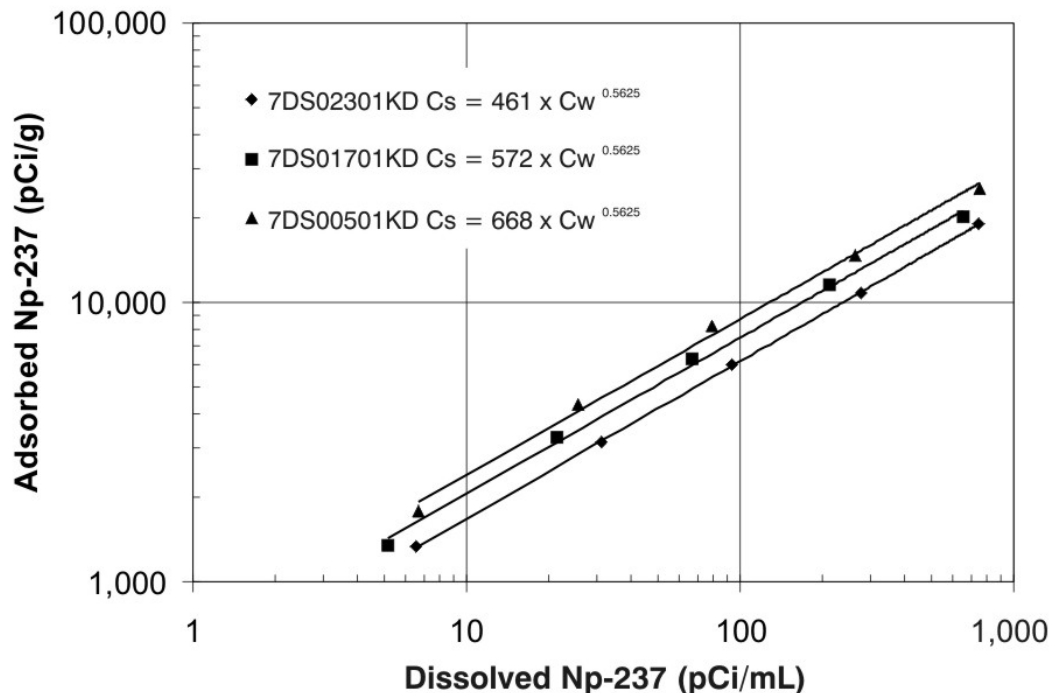
The  $K_f$  values of the Freundlich isotherm showed a large amount of variation, but exponents of isotherms  $n$  were very similar for all samples (see Table 3-41). When plotted on a log-log graph, the Freundlich isotherm becomes linear, with  $n$  being the slope of the line. Parallel lines on the log-log plot indicate how similar the Freundlich  $n$  values are (see Figure 3-48).

Table 3-41. Measured Freundlich and linear isotherm parameters for neptunium and uranium.

Sample ID	Neptunium			Uranium		
	Neptunium Freundlich K	Neptunium Freundlich n	Neptunium $K_d$ (mL/g)	Uranium Freundlich K	Uranium Freundlich n	Uranium $K_d$ (mL/g)
7DS00101KD	246	0.59	100	74	0.81	37
7DS00301KD	91	0.62	27	53	0.78	21
7DS00901KD	100	0.59	25	17	0.78	6
7DS00701KD	24	0.65	6	46	0.76	16
7DS00501KD	155	0.56	37	37	0.81	17
7DS01701KD	133	0.56	34	40	0.77	14
7DS02301KD	109	0.56	28	35	0.81	15
I2S-INEEL-105	52	0.65	15	40	0.78	15
I1S-INEEL-109	280	0.58	98	35	0.78	13
I4D-INEEL-234	86	0.54	16	23	0.76	7
I4D-INEEL-231	124	0.53	27	33	0.76	11
I1D-INEEL-234	95	0.60	25	40	0.87	22
I4D-INEEL-224	162	0.54	41	31	0.81	13
I3D-INEEL-229	256	0.57	85	27	0.77	9
<b>Average</b>	<b>137</b>	<b>0.58</b>	<b>40</b>	<b>38</b>	<b>0.79</b>	<b>16</b>

Note: Information in this table is from Grossman et al. (2001).





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Figure 3-48. Laboratory neptunium adsorption data for three soil samples showing similar slopes when fit with a Freundlich isotherm.

Most of what is known about actinide transport parameters applicable to RWMC comes from 8 years of work at Clemson University and the INL Site (Newman et al. 1996; Fjeld, Coates, and Elzerman 2000; Grossman et al. 2001). Based on these experiments, transport of uranium and neptunium occurs in the dissolved phase in the vadose zone and can be represented by an adsorption type of conceptual model. Uranium, in particular, demonstrates classic breakthrough curves for column tests, suggesting a reversible adsorption mechanism. Neptunium in natural waters is present in the pentavalent(V) valence state (Seaborg and Loveland 1990). An adsorption model for neptunium can be shown to be conservative because at least some of the neptunium is irreversibly adsorbed in column elution experiments. Experimental results from Clemson suggest that reduction of some of the neptunium(V) to neptunium(IV) causes the adsorption. Test results from Clemson indicate that adsorption isotherms for uranium and neptunium are nonlinear. As the uranium and neptunium concentration in the solution increases, the partitioning ratio decreases. Sorption will not be as efficient for retarding movement of uranium and neptunium at higher concentrations as it is at lower concentrations. A Freundlich nonlinear adsorption isotherm would, therefore, be a better approach for uranium and neptunium sorption than a constant  $K_d$  approach.

Plutonium(VI) is not stable in natural waters (Seaborg and Loveland 1990; Choppin, Bond, and Hromadka 1997; Runde 2000; Skipperud, Oughton, and Salbu 2000). Plutonium(V) is the stable oxidation state of dissolved plutonium in natural systems; however, plutonium(V) is reduced to plutonium(IV) through interaction with solid minerals and soil organic matter in natural systems (Skipperud, Oughton, and Salbu 2000). Based on findings of the Clemson batch experiments and on literature review, the batch adsorption experiments at high water-to-solid ratios, using plutonium(VI) as a starting material, failed to be representative of conditions expected in the subsurface at RWMC. Some experiments where plutonium  $K_d$  values are measured using plutonium(V) as a starting material, and at low water-to-solid ratios, are more representative of the vadose zone environment. Under those conditions, a plutonium  $K_d$  over 5,000 mL/g is indicated.

For americium and plutonium, however, transport in the dissolved phase may not be an important mechanism. Attempts to measure adsorption isotherms for americium and plutonium were unsuccessful because americium and plutonium formed insoluble mineral precipitates and could not be kept in solution in the simulated groundwater. This result is consistent with the solubility of solid plutonium and americium minerals in the synthetic water composition. Without a dissolved aqueous phase, a liquid-solid partitioning coefficient could not be measured. A better conceptual model for americium and plutonium seems to be the formation of an insoluble solid phase that is mostly filtered out of solution during batch adsorption experiments, which gives the appearance of a large  $K_d$  value in batch experiments. In column experiments, the particulate material is mostly filtered out of solution by the sediment in the column, also giving the appearance of a large  $K_d$  value for more than 98% of the plutonium.

In column experiments, however, a small but continuous release of trivalent and quatravalent actinides (i.e., thorium, plutonium, and americium) occurs in a particulate form that migrates as fast or almost as fast as the water. This was seen in earlier column experiments conducted by Miner, Evans, and Polzer (1982) and in two generations of studies at Clemson University (Newman et al. 1996; Fjeld, Coates, and Elzerman 2000). A continuous, slow release of a very small fraction of the actinide occurs over time. Even in packed sediment columns, where material had been sieved to sizes less than 0.25 mm, measurable transport of particulate actinides is evident. Because of this characteristic, a tiny fraction of trivalent and quatravalent actinides could be moving with little or no retardation through the vadose zone.

A final factor affecting mobility of actinides identified by the Clemson University studies (Fjeld, Coates, and Elzerman 2000) is the effect of organic complexing agents. Laboratory tests using EDTA, an organic complexing agent, showed that EDTA could greatly enhance actinide mobility. An inventory of complexing agents (e.g., EDTA) used at Rocky Flats Plant (INEEL 1998) indicates that organic complexing agents are likely to be present in the waste. The quantities of EDTA from Rocky Flats Plant are reported as unknown, but a reasonable upper limit of 7.1E+04 kg is suggested in the Historical Data Task report (INEL 1995a, Table 4-1).

### **3.8 Analysis and Leaching of Pit 9 Waste Samples**

The limited retrieval demonstration by the Operable Unit 7-10 Glovebox Excavator Method Project in Pit 9 (see Sections 3.1.5.6) provided samples of retrieved material to the Retrieved Waste and Soil Characterization Project under Operable Unit 7-13/14. Samples of retrieved materials were collected for various purposes (e.g., characterization of waste for compliance with storage or future disposal requirements). Samples (of both soil and waste) were collected intentionally for the Retrieved Waste and Soil Characterization Project to maximize the probability of acquiring highly contaminated material. Groenewold et al. (2005) analyzed and studied actinide leaching of waste and soil in samples collected. Results are summarized in subsections that follow.

#### **3.8.1 Sample Collection Methods**

A total of 44 samples (250 mL each) of soil and waste excavated from Pit 9 were visually categorized, analyzed for total actinide content, evaluated for actinide solubilization, and tested for operational speciation (Groenewold et al. 2005). Samples were initially grouped into three categories: interstitial soil (36 samples), organic sludge (three samples), and inorganic sludge (five samples). However, photographic inspection and chemical analyses showed that the samples were significantly more diverse than indicated by initial categorizations. Soil samples were subcategorized into clean, mostly clean, mixed soil waste, and soil scraped from graphite mold fragments. A fifth category was created when intentional rupture of a jar containing graphite scarfings during excavation resulted in high actinide contamination of visually clean samples in the excavation area.

The three samples originally categorized as organic sludge had chemical characteristics that in many cases would have caused separate consideration; however, the group shared some visual and physical similarities, and the original grouping was maintained. The five samples initially grouped and identified as inorganic sludge were subsequently judged not to be inorganic sludge in the context of material expected from Rocky Flats Plant<sup>i</sup> according to Vejvoda (2005). Chemical analyses suggested that these could be grouped into two categories: Inorganic Waste Type I and Inorganic Waste Type II.

Because Pit 9 was excavated with a backhoe and radioactive particulates were contained by an enclosure, waste material buried in this highly heterogeneous site was mixed before samples could be collected. Groenewold et al. (2005) caution that this method of excavation must affect conclusions that can be drawn about the mechanism of contamination. Because the degree of mixing of waste and interstitial soil cannot be ascertained, attribution of shifts in measured isotopic abundances to contaminant migration could not be rigorously supported.

### 3.8.2 Actinide Contamination

Four soil samples were identified as “clean” (i.e., free of radiological contamination), based on their appearance and actinide concentrations that were either less than detection limits, or in the case of uranium, close to background levels and isotopic distributions expected in nature. The exception to this generalization was presence of very low levels of Pu-240 just above background levels in three of the four samples. The error bars ( $1\sigma$ ) overlap detection limits for Pu-240, underscoring the tenuous nature of these measurements for any one of these samples; yet the fact that Pu-240 was observed repeatedly in three of the four “clean” samples and all other soil samples, but not observed in any of the benchmark samples, suggests that the measurements are real. This low-level contamination notwithstanding, these “clean” or “very nearly clean” soil samples suggest that samples uncontaminated by gross mixing from excavation were acquired.

Thirteen soil samples were categorized as “mostly clean” on the basis of appearance; however, consideration of actinide concentrations suggests that a “low contamination” designation is more appropriate. Eleven of the 13 had Pu-239 concentrations above detection limits and (with one exception) ranged from not detected to about 5.6 nCi/g (90 ng/g). The exception had a Pu-239 concentration of 21 nCi/g (340 ng/g) and probably should have been categorized as a mixed soil waste sample. Plutonium-240 was detected in all 13 samples, and Am-241 was above detection limits in nine of the 13 samples. The uranium concentration was slightly elevated compared to natural background concentration in about half of these samples, and a slight decrease in the U-238/U-235 ratio was measured, consistent with contamination with waste containing enriched uranium.

The Pu-239/Pu-240 mole ratios for the low-contamination soil samples ranged from 16 to greater than 50; these ratios are consistent with weapons-grade plutonium contamination from Rocky Flats Plant process waste. The Pu-239/Am-241 mole ratio was more diagnostic; although the 100-to-500 range is quite broad, it is consistent with those measured for soil scraped from graphite and soil contaminated from the intentional rupture of the jar of graphite scarfings. Contamination in both of these soil sample groups originated from the Rocky Flats Plant process waste. The Pu-239/Am-241 mole ratios were distinct from those measured for waste material. Note that in this study, Am-241 was distinguished from

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i. Because the definition of TRU waste changed in 1982, it is important to note that a large portion of the waste previously designated TRU is not TRU by current standards. Originally, TRU waste was defined as all waste contaminated with TRU radionuclides in concentrations greater than 10 nCi/g (AEC 1973). However, in 1983, TRU waste was redefined based on a concentration of 100 nCi/g (DOE O 5820.1). Currently, TRU waste is defined as waste material containing any alpha-emitting radionuclide with an atomic number greater than 92, a half-life longer than 20 years, and a concentration greater than 100 nCi/g at the end of the period of institutional control as defined in DOE M 435.1-1.

Pu-241; however, because from 76 to 90% of the Pu-241 ( $t = 30$  to 50 years) has already decayed to Am-241, all isotopes with mass 241 were assumed to be Am-241.

A correlation between Pu-239 and Am-241 established for high-plutonium soil samples could be extrapolated to low-concentration soil (see Figure 3-49A, diamond data points in left-hand oval), which suggested that contamination had a similar process origin. In contrast, the Pu-239/Am-241 mole ratios of the low-contamination and high-plutonium soil samples were dissimilar to those measured for waste or soil mixed with waste. This difference is underscored by a plot of the Pu-239/Am-241 mole ratio versus the Am-241 concentration (see Figure 3-49B), which shows that high americium concentration does not correlate with high plutonium in the samples containing waste.

Nine of the interstitial soil samples were categorized as mixed soil waste. Actinide concentrations for these samples displayed significantly greater variability. For example, Pu-239 ranged from not detected in one instance to 43 nCi/g (700 ng/g). Elevated uranium (with a depressed U-238/U-235 mole ratio), Pu-240, and Am-241 were measured in each of these samples. The Pu-239/Pu-240 mole ratio ranged from 12 to 20, consistent with ratios expected from weapons-grade plutonium. The Pu-239/Am-241 ratio, on the other hand, had more of a bimodal distribution: five of the samples had ratios ranging from 90 to 190, consistent with Rocky Flats Plant scarfings, as noted previously for the low-contamination soil samples. However, three samples had Pu-239/Am-241 mole ratios less than 10, which was more in accord with that measured for five of the eight waste samples. This suggests a different process origin for contamination in this material.

One soil sample was collected by scraping off soil adhering to fragments of a graphite mold directly into a sample container. This occurred before the intentional rupture of the jar of graphite scarfings; therefore, this sample is a valuable benchmark of contaminated soil found close to a graphite fragment. The Pu-239 concentrations measured for multiple subsamples were exceptionally high and were highly variable, ranging from 1,900 to 4,800 nCi/g (31,000 to 78,000 ng/g). The sample did not contain elevated U-238, but did contain significant U-235, U-236, Np-237, Pu-240, Am-241, and Pu-242. However, Pu-239 was by far the most abundant actinide isotope: Pu-239/Pu-240 ratios were consistently about 16, in agreement with Rocky Flats Plant plutonium isotopic distributions. The Pu-239/Am-241 mole ratios were about 200 in this sample.

Nine soil samples with a clean appearance were collected after the intentional rupture of the jar of graphite scarfings; in most respects, these samples were analytically indistinguishable from the soil scraped from the graphite. Concentrations of 1,860 to 3,900 nCi/g (30,000 to 63,000 ng/g) were measured for Pu-239, and similar results were recorded for other isotope concentrations and ratios measured for the sample of soil scraped from graphite. This suggests that contamination both in the soil scraped from graphite and in the soil after rupture of the jar of graphite scarfings may have similar origins.

Actinide content of the samples identified as waste also varied greatly. All three of the organic waste samples had significantly elevated concentrations of the isotopes of interest (Np-237 was not detected in any of them), with Pu-239 at nearly 310 nCi/g (5,000 ng/g) for one sample. For two of the organic sludge samples, the Pu-239/Pu-240 mole ratio was about 12, indicating that this was not entirely weapons-grade plutonium. This view was supported by very low Pu-239/Am-241 mole ratios (about 2) for these samples, suggesting that these samples were derived from other processes. The third organic sludge had Pu-239/Pu-240 and Pu-239/Am-241 mole ratios consistent with the soil scraped from graphite, although plutonium concentrations in this sample were much lower.

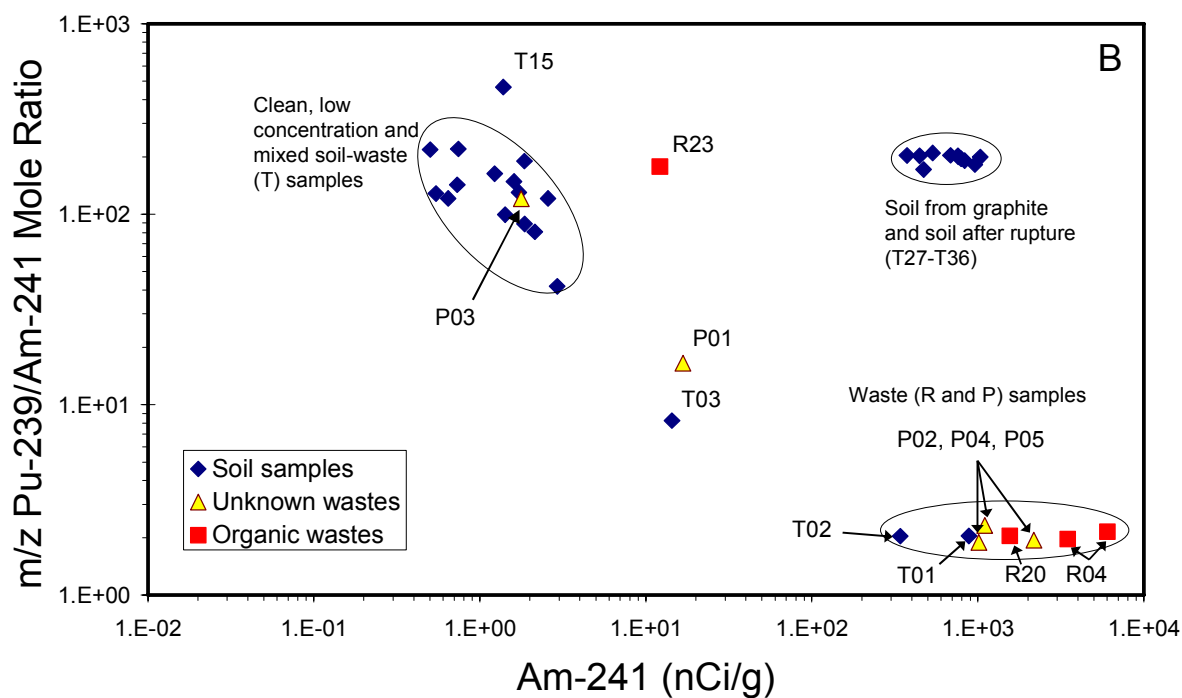
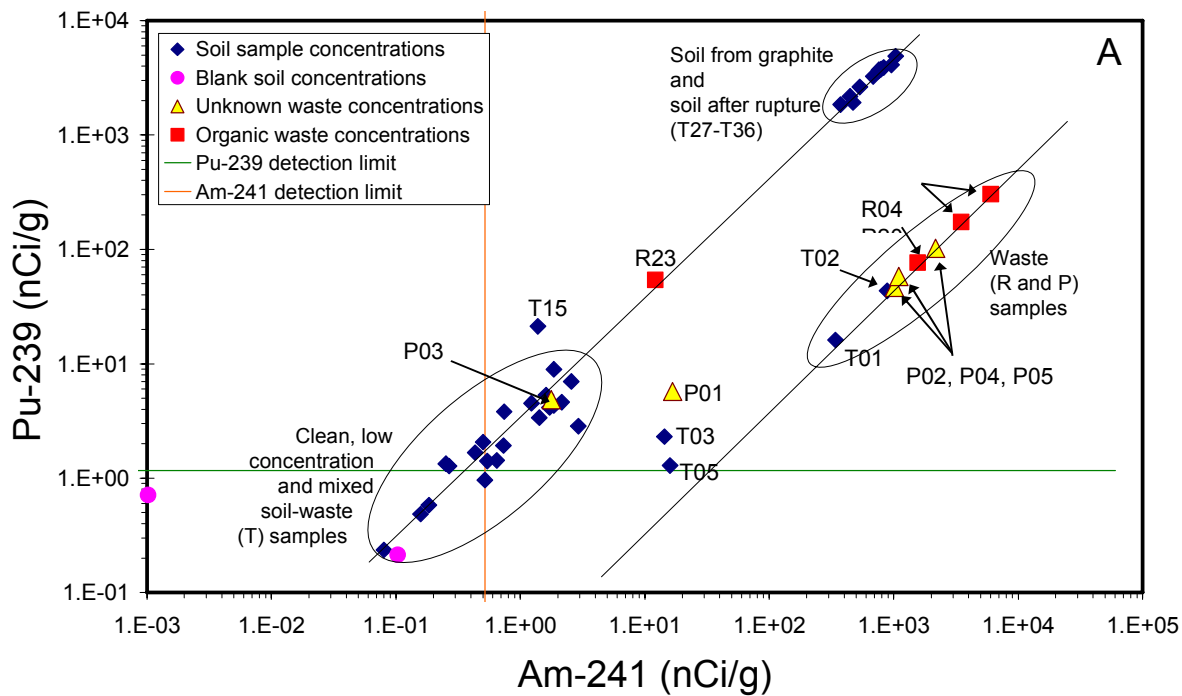


Figure 3-49. Plots of (A) Pu-239 versus Am-241 and (B) the Pu-239/Am-241 isotope ratio versus Am-241.

Three of the waste samples originally categorized as inorganic waste had Pu-239 concentrations and Pu-239/Pu-240 and Pu-239/Am-241 mole ratios that were very similar to those of the two high-plutonium organic waste samples, despite their physical appearance being markedly different. A fourth unknown waste sample had actinide concentrations and isotope ratios that were very similar to the low-concentration interstitial soil samples. The fifth unknown waste sample had low but significant concentrations of Pu-239 and Am-241, but displayed isotope ratios that were unlike any of the other categories. This again emphasizes the variability that can be encountered.

### 3.8.3 Aqueous Partitioning of Actinide Contaminants

Phenomenological distribution coefficients ( $K_d$ ) were measured for actinide isotopes in a subset of samples under conditions of varying pH and ionic strength. These studies provided a quantitative assessment of actinide dissolution from waste material and subsequent adsorption to the adjacent soil. Commonalities observed by Groenewold et al. (2005) among the different actinide elements are listed below:

- $K_d$  values decrease significantly as the pH of the leachate solution becomes acidic
- $K_d$  values for organic waste samples are much more sensitive to decreased pH than are soil samples
- In the pH range typical of INL Site soil (about 8),  $K_d$  values are not significantly affected by small changes in pH
- At high pH ranges,  $K_d$  values frequently undergo modest decreases, thought to be due to dissolution of the matrix.

**3.8.3.1 Uranium Partitioning.** Uranium partitioning displayed complex behavior.  $K_d$  values of  $10^3$  to  $10^4$  mL/g were measured in the pH 8-to-12 range for all samples. Below pH 8,  $K_d$  values dropped sharply, reaching local minima at about pH 6 that were one to two orders of magnitude lower than those at high pH. The  $K_d$  minimum at pH 6 indicates a significant alteration in the chemistry of the system, which is reversed at mildly acidic pH values where  $K_d$  values once again rise. Increases in partitioning coefficients in the pH 4-to-5 range are dramatic (to about  $10^4$  mL/g) for soil samples, but are more modest for waste samples. Finally, at very acidic pH regimes,  $K_d$  drops to between  $10^1$  and  $10^2$  mL/g for the soil samples, where uranium is essentially dissolved from waste samples. Generally, ionic strength had little effect on  $K_d$  values for uranium.

The partitioning of anthropogenic uranium (e.g., waste contaminated with U-235 enriched) at high pH is somewhat greater than that of naturally occurring uranium. This conclusion is based on the observation that the U-238/U-235 ratio in the leachate dropped regularly as pH increased.

**3.8.3.2 Plutonium Partitioning.** In contrast to uranium, plutonium partitioning varies more regularly and depends greatly on the solid matrix (see Figure 3-50). For organic waste samples exposed to pH less than 3,  $K_d$  values as low as the mid- $10^1$ -mL/g range are measured. The  $K_d$  rises to  $10^3$  mL/g at pH 3 to 7, and then rises to the  $10^5$ -mL/g range at higher pH. For low-contamination soil samples, plutonium  $K_d$  values as low as  $10^3$  mL/g are measured at pH 1, but rise steadily to about  $10^4$  mL/g by pH 3, and hover between  $2 \times 10^3$  and  $2 \times 10^4$  mL/g over the remainder of the pH range studied.

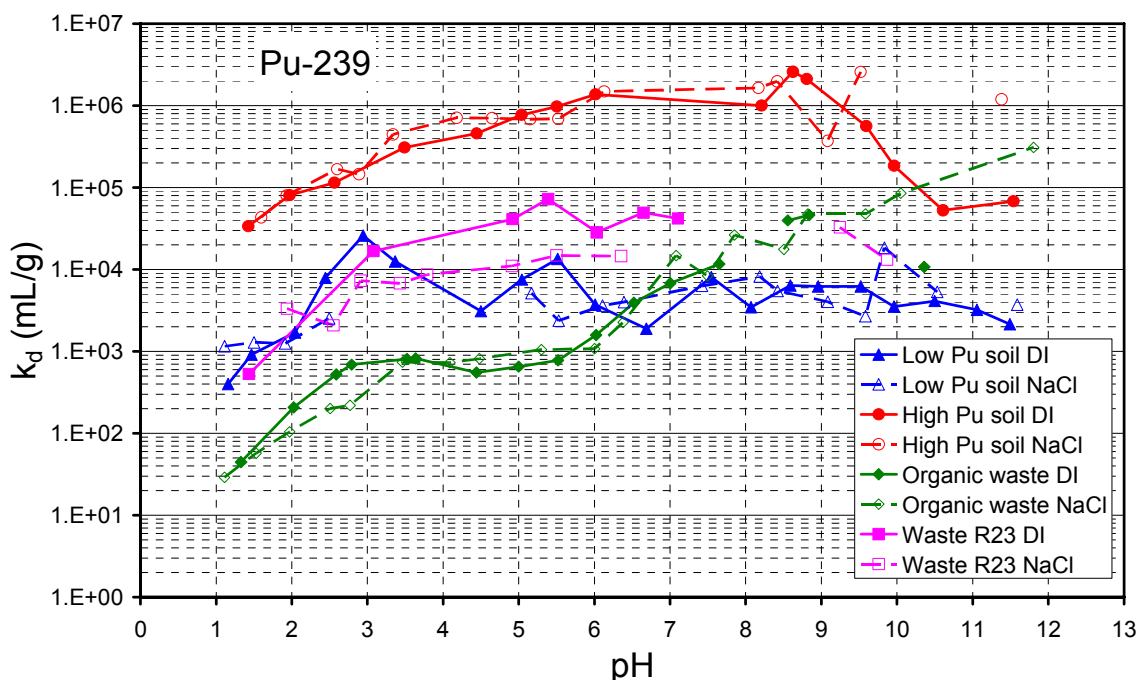


Figure 3-50. Average  $K_d$  values for leaching Pu-239 with low-ionic-strength leachate (deionized water) and high-ionic-strength leachate (100 mm sodium chloride) as a function of pH for interstitial soil samples and organic waste.

Plutonium partitioning is markedly lower in soil scraped from graphite and in soil acquired after the intentional rupture of the graphite jar. At low pH,  $K_d$  values in the mid  $10^4$  mL/g range are measured; however, as pH rises to ambient pH levels (i.e., 5 to 10),  $K_d$  for plutonium increased to  $10^6$  mL/g. At the highest pH ranges, a modest decrease in  $K_d$  is observed, and this may signal some dissolution of the silicate matrix.

**3.8.3.3 Americium and Neptunium Partitioning.** Americium partitioning in the ambient-to-high pH range (about 6 to 12) is described by  $K_d$  values that range from  $10^4$  to  $10^5$  mL/g; low-contamination soil samples display somewhat lower values at very high pH due to dissolution of the silicate matrix. Beginning at pH 6,  $K_d$  for soil samples begins to decrease as the pH is lowered, finally reaching about  $10^2$  mL/g at pH 2. The decrease with decreasing pH is even sharper for organic sludge samples. A  $K_d$  less than  $10^1$  mL/g was recorded at pH values just slightly less than 4 for organic waste samples.

Neptunium partitioning from soil samples was characterized by a  $K_d$  of about  $10^2$  mL/g at low pH, rising to about  $10^3$  mL/g as pH increases to about 7. Similar values were measured for the waste samples at high pH; however, in these samples,  $K_d$  drops sharply with decreasing pH, reaching a value of less than  $10^1$  mL/g by pH 5, and probably achieving complete dissolution at lower pH values.

### 3.8.4 Operational Speciation

Studies using sequential aqueous extraction showed susceptibility of actinide elements to dissolve under perturbed chemical conditions and provided qualitative insight into operational speciation within the solid sample matrix. Uranium in soil samples tended to reside predominantly in the residual fraction (i.e., it would not leach under operational conditions imposed by sequential aqueous extraction). Depending on the particular sample being studied, smaller percentages of uranium were partitioned into



carbonate, oxidizable, and reducible fractions, with oxidizable being second in importance to residual. These tendencies were even more pronounced in the high-plutonium soil samples from the graphite scarfings, in which bias toward the residual fraction was even stronger. Uranium in organic waste samples tended to be more evenly distributed among the residual, oxidizable, and carbonate fractions, which reflects more heterogeneous speciation and less aggressive binding of this matrix.

Plutonium in low-concentration soil samples tends to be fairly evenly partitioned between the oxidizable and reducible fractions. Speciation of plutonium in the oxidizable fraction is probably due to oxidizable plutonium that is present as insoluble Pu(IV) oxides or hydroxides that are susceptible to oxidation, forming more soluble Pu(V) or Pu(VI) species, or perhaps to destruction of matrix binding sites upon oxidation. This conclusion is supported by the fact that the largest percentages of plutonium were measured in the oxidizable fraction from the sequential aqueous extraction of plutonium-spiked soil that contained almost no organic matter. Plutonium in reducible fractions is commonly attributed to metal bound with iron sesquioxide minerals; these are certainly present on the surfaces of silicate-soil particles or as rusted-barrel debris within the burial site.

For the highly contaminated samples scraped from graphite or contaminated by the intentional rupture of the graphite jar, the plutonium distribution shifted strongly toward the residual fraction. This shift is consistent with the very high  $K_d$  values measured for these samples. Similar patterns were observed for the waste samples; plutonium tended to reside in the residual or nonleachable fraction.

Operational speciation of americium was studied only in soil samples that were highly contaminated with plutonium and in organic waste samples. A significant difference was observed when americium partitioning in the soil scraped from graphite was contrasted with the soil after intentional rupture of the jar of graphite scarfings. In the soil scraped from graphite, the largest percentage of americium was in the reducible fraction, and significant percentages also were in the oxidizable and carbonate fractions. This was similar to americium behavior in the plutonium-spiked soil samples. In sharp contrast, americium in soil samples contaminated by the rupture resided almost exclusively in the residual fraction, again suggesting that americium is unlikely to leach from this material. Similarly, the residual fraction also contained the largest percentage of americium in the organic sludge samples.

Neptunium operational speciation could only be assessed for two samples: the soil scraped from graphite and one organic sludge. In both instances, the oxidizable fraction contained the largest percentage of neptunium, with the carbonate also significant. This suggests that sorbed neptunium may be present in a reduced form.

### **3.9 Carbon-14 and Tritium from Activated Beryllium**

Modeling of release and migration of C-14 for Operable Unit 7-13/14 has been improved by using multiple-phase reactive-transport approaches, bench-scale and near-field-scale experiments, and over 5 years of field data to use for model calibration. Recent modeling to estimate quantities and concentrations of radioactivity in the beryllium blocks, monitoring of beryllium disposals in SVR 20, and activated nonberyllium metal disposals in SVR 12 are summarized in the following subsections. This section also briefly describes experiments and models used to explore the fate of C-14 and tritium released from activated metal disposals in the SDA.

### 3.9.1 Estimated Activity in Beryllium Blocks Buried in the Subsurface Disposal Area

Research reactors at RTC, including Materials Testing Reactor, Engineering Test Reactor, and Advanced Test Reactor, use beryllium reflectors. Beryllium is used as a neutron reflector to intensify the neutron flux in the reactor core. The amount of beryllium used varies in each reactor (Sebo et al. 2005). The Advanced Test Reactor reflector assembly consists of a set of eight beryllium blocks and 16 outer shim control cylinders (see Figure 3-51). Each block is 129.5 cm (51 in.) long, approximately 40.6 cm (16 in.) square, and weighs 81.42 g (179.5 lb). When all eight blocks are assembled, they have a cross section approximately 127 cm (50 in.) in diameter. For the Engineering Test Reactor, the reflector assembly was essentially four slabs that surrounded the core, while the Materials Testing Reactor reflector assembly was much more complex.

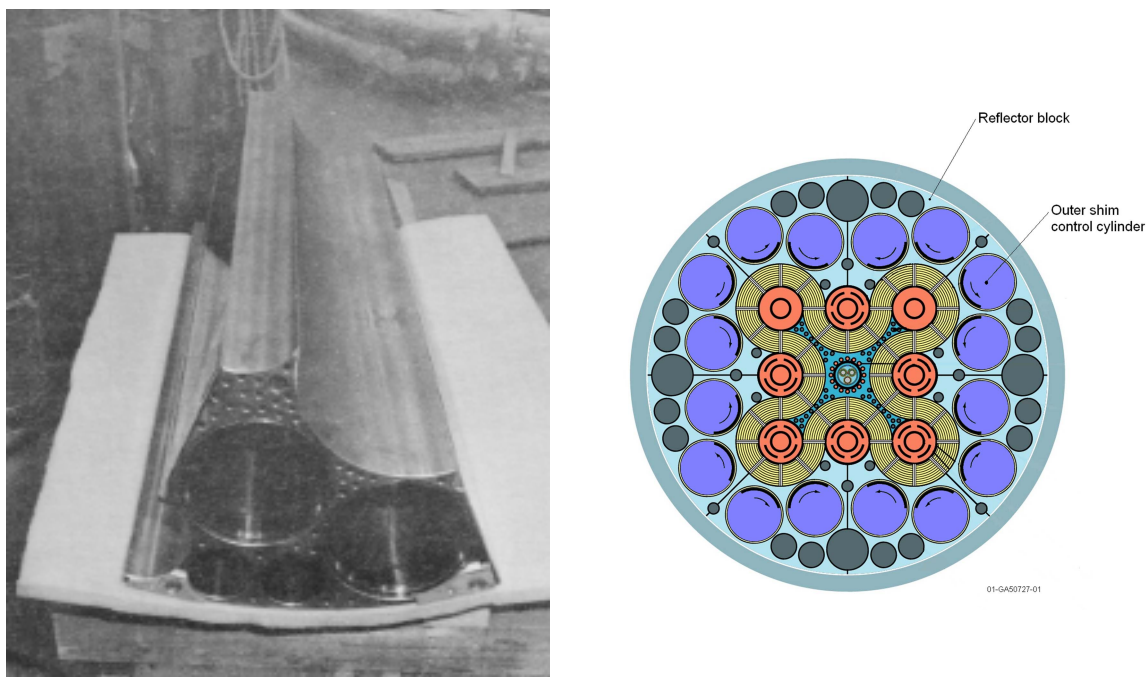


Figure 3-51. Advanced Test Reactor beryllium reflector block.

Historically, reflector assemblies from test reactors periodically were replaced every 8 to 10 years because of swelling. The majority of irradiated beryllium reflector waste was buried in the SDA in three major events: 1976, 1977, and 1993.<sup>j</sup> A total of 5,309 kg (11,703 lb) of beryllium from the Advanced Test Reactor, Engineering Test Reactor, and Materials Testing Reactor (see Table 3-42) (Sebo et al. 2005) was buried in the SDA. Early efforts to characterize the beryllium blocks relied on modeling alone to develop isotope inventories; these efforts could have overestimated certain key nuclides, which was a concern. To reduce conservatism in inventory estimates, samples were taken from stored irradiated beryllium blocks at RTC. Analysis was necessary to determine C-14 content. Limitations on C-14 content for LLW destined for disposal in the SDA are defined in the INL Waste Acceptance Criteria (DOE-ID 2005a), based on the LLW operation performance assessments (Maheras et al. 1994; Case et al. 2000). Similar analysis had not been required for previous beryllium disposals because earlier versions of the waste acceptance criteria did not limit C-14.

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j. Most beryllium blocks in the SDA were grouted in a non-time-critical removal action in 2004. See Section 3.1.6 for a description of methods, coverage, and efficacy of beryllium encapsulation.

Sampling showed that the initial models were indeed conservative with regard to C-14, but Nb-94 was substantially underestimated (Sebo et al. 2005). Sample results also revealed that trace impurities in the beryllium blocks, when subjected to neutron flux in a reactor, transmute to other elements. Uranium is transmuted into plutonium and other TRU isotopes, and gold is transmuted into mercury. Modeling based on reactor burn-up histories and estimates of concentrations of impurities in the beryllium has shown that the irradiated beryllium blocks contain sufficient TRU content to be classified as TRU waste. Table 3-42 provides estimates of C-14 and TRU concentrations in beryllium blocks buried in the SDA. Figure 3-52 illustrates the locations of the beryllium blocks in the SDA.

### 3.9.2 Monitoring Beryllium Reflector Blocks

Six beryllium reflector blocks from Advanced Test Reactor were buried in SVR 20 in 1993 (see Figure 3-52) (Ritter and McElroy 1999). The blocks contained a total of 293,000 Ci of tritiated hydrogen gas and approximately 20 Ci of C-14 at their time of disposal. Both radionuclides form mobile compounds. About one-fifth of the total C-14 inventory in the SDA is associated with beryllium. Carbon-14 was identified as a contaminant of potential concern in the IRA (Becker et al. 1998) and as a risk driver in the RWMC Performance Assessment (Case et al. 2000) and the Composite Analysis (McCarthy et al. 2000). Tritium, though not a risk driver, was identified as a contaminant of interest because of its potential as a model calibration target for vapor-phase transport. Additionally, estimated annual emissions of tritium from buried activated beryllium at SVR 20 are used to develop emission estimates for all buried beryllium as required by the “National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities” (40 CFR 61, Subpart H).

Dedicated monitoring to characterize migration of tritium and C-14 from buried beryllium reflector blocks began in 1994 and continued until September 2003. Monitoring probes in SVR 20 were abandoned to prepare for activities associated with the Operable Unit 7-13/14 Early Actions Beryllium Encapsulation Project (Lopez et al. 2004). Surface recontouring and excavation of the grouting location (see Section 3.1.6) required removing instruments from SVR 20. New vapor probes were subsequently installed in 2004 and will be used for continued monitoring. These probes are located in two bundles at 5.8 and 8.8 m (19 and 29 ft) from beryllium reflector blocks. Probes were installed at a greater distance from the buried beryllium to avoid the grout containment unit and grout-affected soil. Results of environmental monitoring, characterizing releases and environmental conditions around the beryllium blocks in SVR 20, are summarized in the following subsections. Because of the large difference in port locations, data from the new vapor probes are presented separately from those from the abandoned probes.

**3.9.2.1 Airborne Tritiated Water Vapor.** Ambient air samples were collected 30 cm (1 ft), 1 m (3.3 ft), and 2 m (6.6 ft) above ground near SVR 20 (see Figure 3-53) during summer and early fall of 2000 through 2004. Samples were collected weekly because the emission rate can vary substantially over relatively short periods. Passive airborne tritium samplers are used to collect samples. Before 2000, samples were collected by several organizations. A summary of 1995 to 1998 results and a description of sampling methods is provided by Ritter and McElroy (1999, Section 3.1). Results for all beryllium sampling are shown in Figure 3-53 (Parsons, Seitz, and Keck 2005). Typically, the concentration of tritiated water vapor in air ranged over several orders of magnitude each year; concentrations peak in late summer. Usually, maximum concentrations persist over a few weeks each year, but the concentration peaked over a 4-day period during 1997. Estimated annual tritium release to the atmosphere ranged from less than 1 Ci in 1998 to more than 100 Ci in 1995 and 1996. Variation is probably caused by year-to-year differences in soil conditions that affect gas-phase permeability and by possible changes in rate of tritium release from the blocks by corrosion. Monitoring data do not show any long-term trend in air concentration or emission, though seasonal variability of the emission rate is evident.

Table 3-42. Summary of the Advanced Test Reactor, Engineering Test Reactor, and Materials Testing Reactor irradiated beryllium reflector waste buried in the Subsurface Disposal Area.

Initial Irradiated Date	Final Irradiated Date	Core	Beryllium Metal Mass (g)	Metal Volume (m <sup>3</sup> )	Disposal Date	Disposal Location <sup>a</sup>	Total C-14 (Ci)	C-14 Concentration (Ci/m <sup>3</sup> )	Transuranic Concentration (nCi/g)	Total Transuranic Inventory (Ci)
3/31/52	7/3/69	MTR 1	~616,000	~0.333	1970	Trench 52	9.01	27.1	694	0.426
3/31/52	7/3/69	MTR 1	~1,384,000	~0.747	1977	Trench 58	20.2	27.0	854	1.18
10/15/57	3/1/70	ETR 1	~624,000	~0.337	1970	Trench 54	21.7	64.4	257	0.160
2/1/68	9/9/72	ATR 1	651,360	0.352	1976	Trench 58	7.81	22.2	319	0.210
2/5/73	4/11/77	ATR 2	488,520	0.264	1982	SVR 12	5.83	22.0	327	0.160
8/9/77	2/2/86	ATR 3	488,520	0.264	1993	SVR 20	12.0	45.3	379	0.185
2/5/77	4/11/77	ATR 1 and 2	489,881	0.2648	1987	SVR 17	15.9	60.1	297	0.145
<b>Total</b>			<b>~4,742,300</b>	<b>~2.6</b>			<b>92.5</b>			<b>2.47</b>

Note: Data in this table are from Sebo et al. (2005).

a. A non-time-critical removal action to encapsulate beryllium blocks was deployed in 2004 (see Section 3.1.6). Blocks in SVR 12 were not grouted.

ATR = Advanced Test Reactor

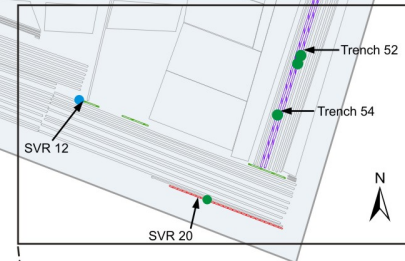
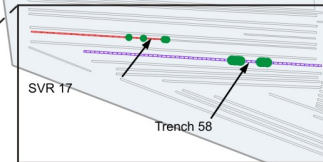
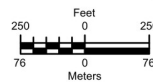
ETR = Engineering Test Reactor

MTR = Materials Testing Reactor

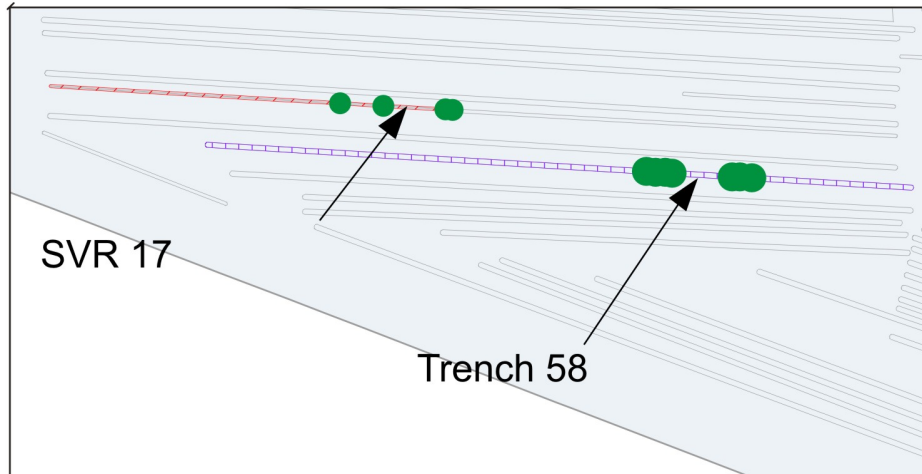
SVR = soil vault row

# Subsurface Disposal Area

- Nongrouted beryllium site
- Approximate locations of grouted beryllium reflector blocks
- SVR 12
- SVR 17 and 20
- Trenches 52, 54, and 58
- Pit, trench, or SVR boundary



## Detail



## Detail

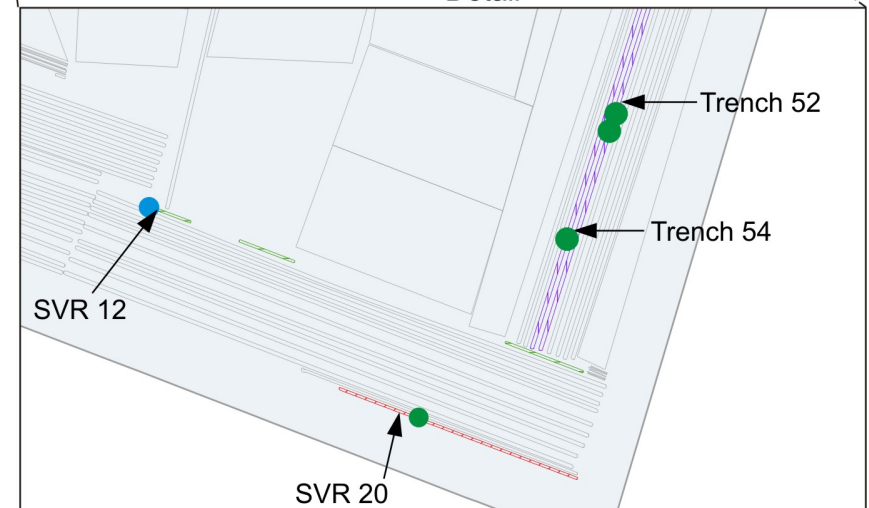


Figure 3-52. Beryllium disposal locations in the Subsurface Disposal Area.

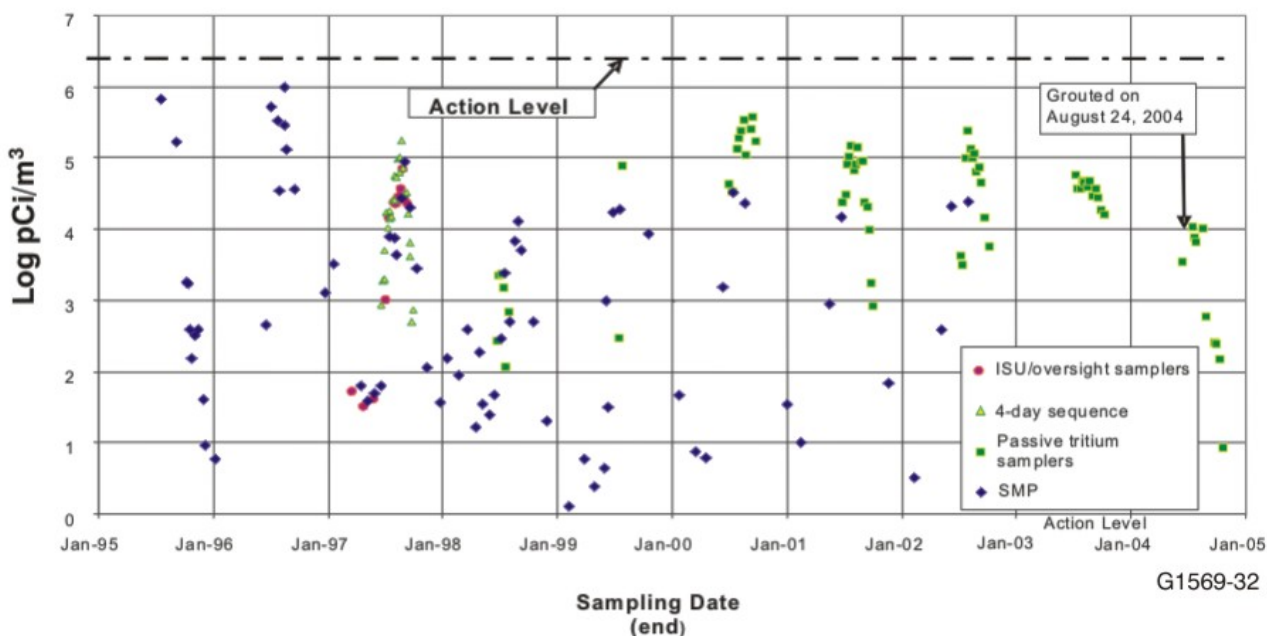


Figure 3-53. Concentrations of airborne tritiated water vapor above beryllium blocks buried in Soil Vault Row 20.

**3.9.2.2 Tritiated Water Vapor in Soil-Gas Samples: 1996 to 2003.** From 1996 to 2003, soil-gas samples were collected approximately 60 to 100 cm (23 to 39 in.) from the buried beryllium blocks in SVR 20 from Gas-sampling Port (GSP) -1. Soil-gas data through September 22, 2003, are summarized in Figures 3-54, 3-55, and 3-56 (Koeppen et al. 2004). Maximum concentrations of tritiated water vapor in soil moisture were 5.8, 0.98, and 0.90  $\mu\text{Ci/mL}$  at depths of 2.7, 4.5, and 6.2 m (9, 15, and 20 ft), respectively. Correlation of the concentrations at 4.5 and 6.2 m (15 and 20 ft) is depicted in Figure 3-57. The concentration of tritiated water vapor in soil moisture appears to fluctuate over the course of each year, with lower concentrations occurring during cool months.

**3.9.2.3 Tritiated Water Vapor in Soil-Gas Samples: 2004 and 2005.** Two soil vapor port bundles were installed to monitor tritium and C-14 in SVR 20 after grout injection. Soil vapor port bundles RWMC-2022 and RWMC-2023 are located 5.8 and 8.8 m (19 and 29 ft), respectively, from activated beryllium disposals in SVR 20. Both vapor probes have ports open at depths of 1.2, 2.7, and 4 m (4, 9, and 13 ft). Because of differences in the depth to the basalt layer, the deepest port on RWMC-2022 is located at 5.9 m (19.5 ft), and the deepest port on RWMC-2023 is located at 5.6 m (18.5 ft). Sampling began in September and December 2004 on RWMC-2022 and -2023, respectively. Results to date have not been published, but will be included in future monitoring reports. Soil-gas data are summarized in Table 3-43. Maximum concentrations from these ports are approximately three orders of magnitude less than those recorded from GSP-1 due to increased distance from the beryllium blocks. Note the four-fold difference between maximum concentrations recorded at the 1.2-m (4-ft) depths of RWMC-2022 and -2023 over a 3-m (9.8-ft) distance. Concentrations at each port decrease with increasing depth, as observed in the results from GSP-1. Concentrations at each port vary over the sampled period. A more extensive data set for these ports is needed before any conclusions may be drawn about the efficacy of grouting in FY 2004.

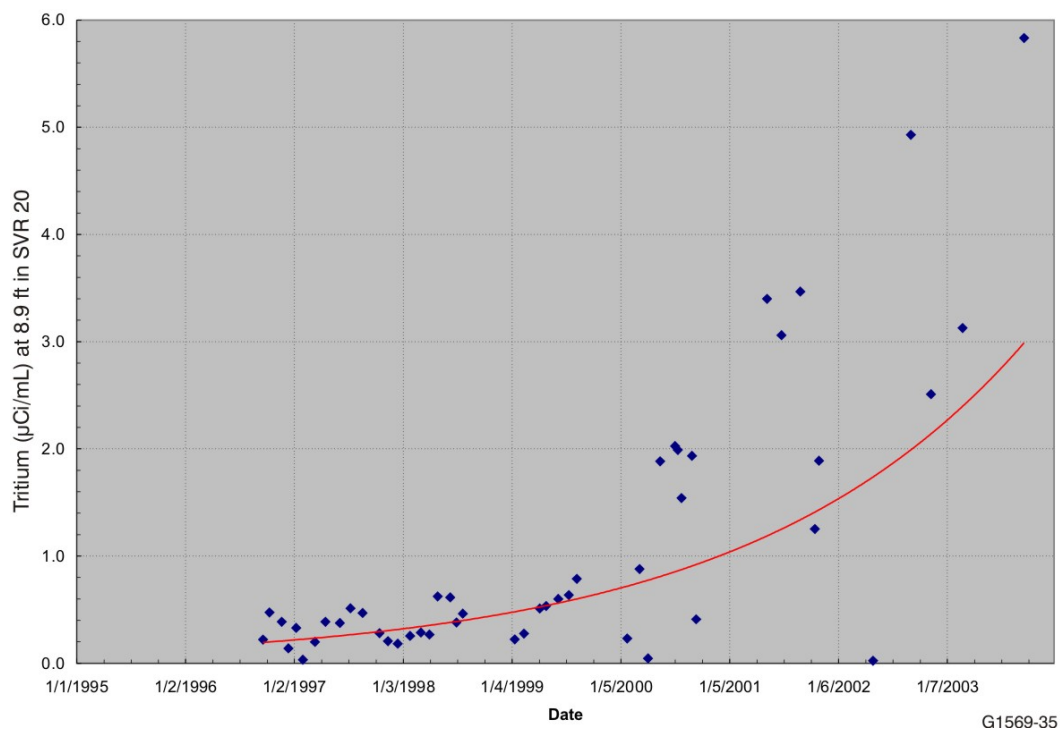


Figure 3-54. Concentrations of tritiated water in soil moisture collected from a depth of 8.9 ft near activated beryllium in Soil Vault Row 20.

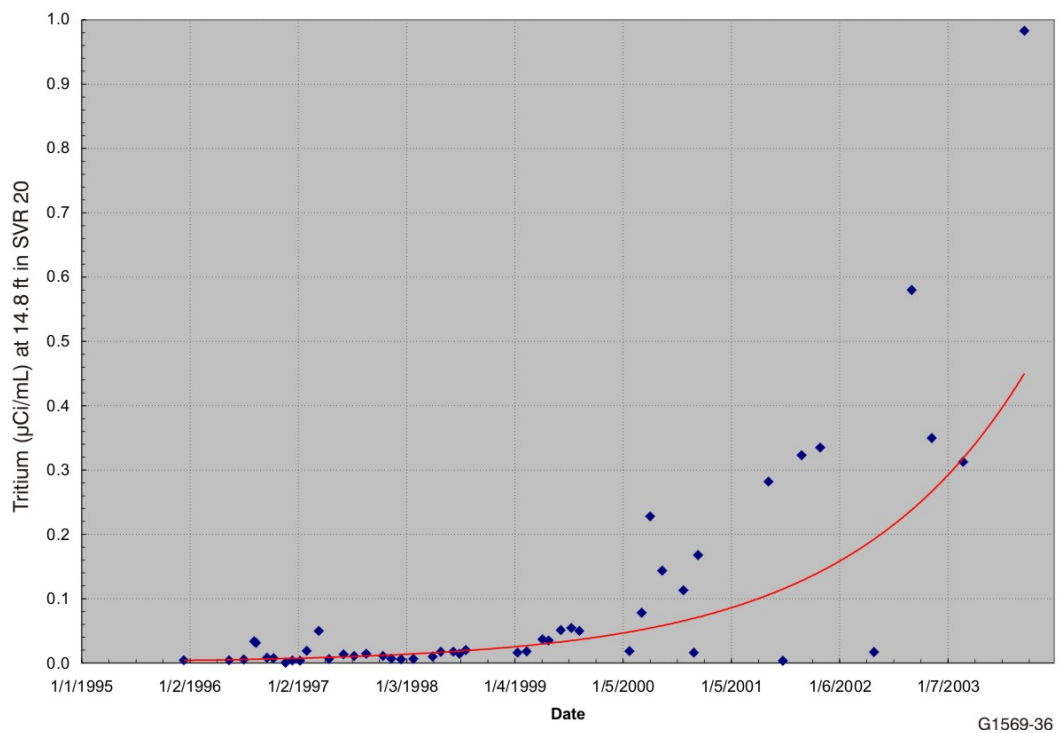


Figure 3-55. Concentrations of tritiated water in soil moisture collected from a depth of 14.8 ft near activated beryllium in Soil Vault Row 20.



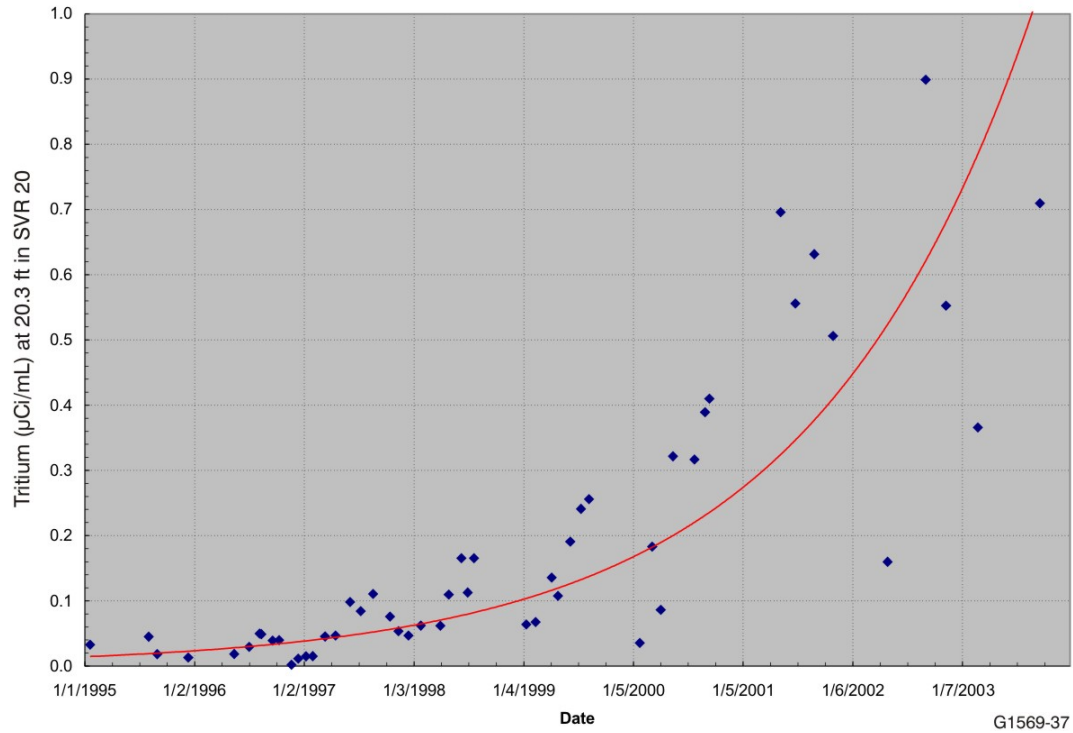


Figure 3-56. Concentrations of tritiated water in soil moisture collected from a depth of 20.3 ft near activated beryllium in Soil Vault Row 20.

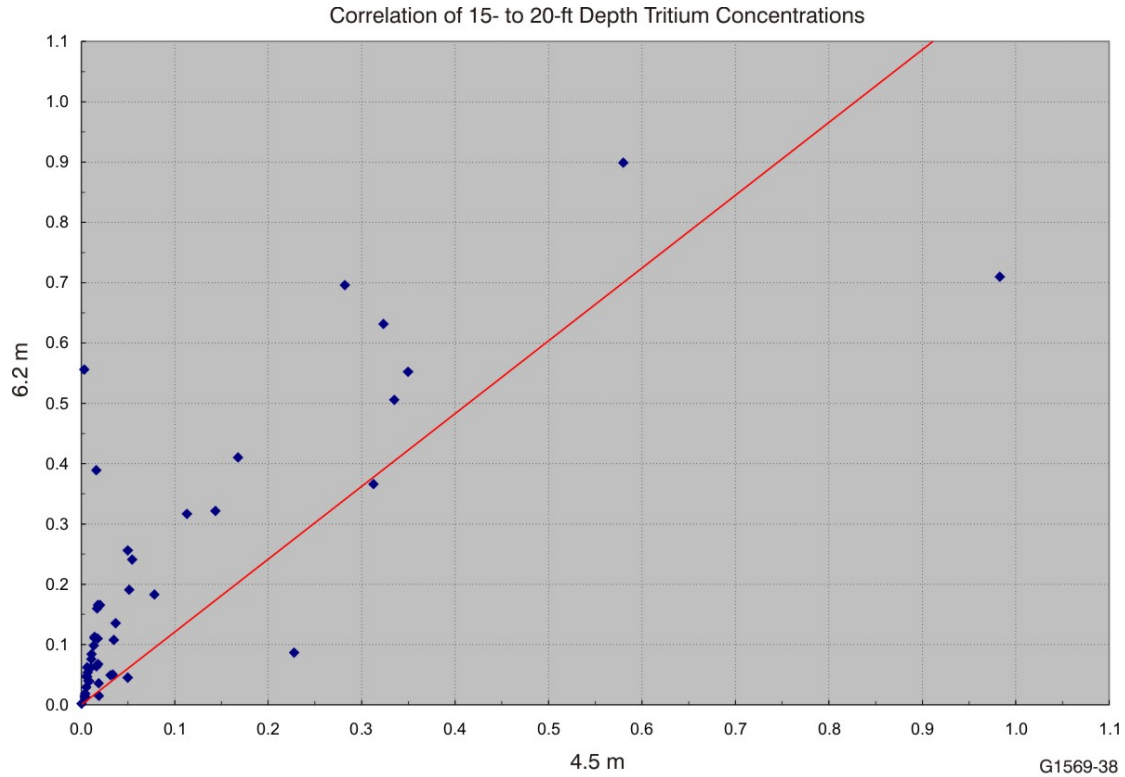


Figure 3-57. Correlation of concentrations of tritiated water in soil moisture collected from depths of 15 and 20 ft in Soil Vault Row 20.



Table 3-43. Summary of tritiated water activity (pCi of tritium per mL water) in soil-gas samples collected from Probes RWMC-2022 and -2023 at Soil Vault Row 20.

Date	RWMC-2022				RWMC-2023			
	Depth				Depth			
	1.2 m	2.7 m	4.6 m	5.9 m	1.2 m	2.7 m	4.6 m	5.6 m
9/29/2004	7,553	106	146	242	—	—	—	—
12/7/2004	8,342	91	155	159	—	—	—	—
12/14/2004	—	—	—	—	1,459	123	98	4
2/21/2005	8,103	94	—	122	—	—	—	—
2/24/2005	—	—	—	—	1,612	112	53	3
5/12/2005	6,323	90	99	104	1,797	46	14	2
6/9/2005	6,355	95	95	103	1,231	46	13	3
7/11/2005	6,992	105	107	105	506	75	17	3

**3.9.2.4 Airborne Carbon-14 Vapor.** Total emissions of C-14 in carbon dioxide from buried activated beryllium have been estimated for the National Emission Standards for Hazardous Air Pollutants annual reports, assuming 100% of the C-14 released to the subsurface migrates to the atmosphere (Olson et al. 2003). These conservative estimates indicate that C-14 emissions do not contribute significantly to off-INL Site dose through the air pathway; therefore, periodic or confirmatory measurements are used to support modeled emission estimates. The first periodic or confirmatory measurement was collected at SVR 20 during FY 2002. The concentration of C-14 in the air, measured in a grab sample, was less than 1 nCi/m<sup>3</sup>—the detection limit for the sampling or analysis method. No subsequent airborne C-14 measurements have been made.

**3.9.2.5 Carbon-14 in Soil Gas.** The specific activity of C-14 in carbon dioxide (i.e., the C-14 activity per gram of total carbon) was measured in grab samples of soil gas collected at various depths from the three GSP-1 ports and Probe SVR20-5-VP3. Samples collected through 1997 used caustic solutions in bubblers; samples collected after 1997 used a Tedlar bag method (Plummer, Hull, and Fox 2004; Fox et al. 2004). The two sampling methods are assumed to produce comparable data. Sample results from 1996 to 2004 are summarized in Table 3-44 (Koeppen et al. 2005). Based on a review of all results for the 1999 to 2002 monitoring, C-14 specific activity (i.e., pCi of C-14 per gram of carbon in carbon dioxide) in soil gas near buried beryllium has increased by a factor of 5 to 10 since the late 1990s. A similar increase in tritiated water vapor concentrations in soil water is evident over the same period. After the removal of GSP-1 and Probe SVR20-5-VP3, samples were collected from RWMC-2022 and -2023 (described in the previous section). Results to date have not been published but will be included in future monitoring reports. Soil-gas data are summarized in Table 3-45. The abandoned Probe SVR20-5-VP3 and existing Probe RWMC-2022 are located approximately 3 to 5 m (9.8 to 16.4 ft) from the activated beryllium disposal and have produced samples with comparable C-14 specific activities, as demonstrated in this limited data set.

Table 3-44. Summary of carbon-14 specific activity (pCi of carbon-14 per gram of carbon) in carbon dioxide in soil-gas samples collected from abandoned probes at Soil Vault Row 20.

Date	GSP-1	GSP-1	GSP-1	SVR20-5-VP3
	Depth 2.7 m	Depth 4.5 m	Depth 6.2 m	Depth 5.4 m
6/5/1996	1.7E+04	—	2.3E+04	—
7/2/1996	—	3.4E+04	2.5E+04	—
12/12/1996	1.3E+05	4.2E+04	3.3E+04	—
11/12/1997	4.4E+04	2.0E+04	1.2E+04	—
11/15/2001	3.6E+05	1.5E+05	2.4E+05	8.2E+04
2/20/2002	—	—	—	2.8E+04
5/2/2002	1.7E+05	1.4E+05	1.3E+05	—
5/23/2002	—	—	—	3.1E+04
8/23/2002	1.3E+05	1.3E+05	1.3E+05	3.6E+04
11/13/02	—	—	—	2.4E+04
11/5/03	1.1E+05	1.4E+05	1.6E+05	4.1E+04
03/30/04	5.8E+04	6.4E+04	9.5E+04	—

Note: Data in this table were taken from Koeppen et al. (2005).

Table 3-45. Summary of carbon-14 specific activity (pCi of carbon-14 per gram of carbon) in carbon dioxide in soil-gas samples collected from probes at Soil Vault Row 20.

Date	RWMC-2022				RWMC-2023			
	Depth				Depth			
	1.2 m	2.7 m	4.6 m	5.9 m	1.2 m	2.7 m	4.6 m	5.6 m
12/7/2004	5.17E+04	4.44E+04	4.30E+04	4.57E+04	—	—	—	—
2/21/2005	4.71E+04	3.86E+04	—	3.11E+04	—	—	—	—
6/9/2005	4.85E+04	4.42E+04	4.09E+04	3.92E+04	1.79E+04	2.05E+04	2.32E+04	2.82E+04

**3.9.2.6 Chloride in Soil Moisture.** Suction lysimeters were installed at depths of 2 and 6 m (7 and 20 ft) near buried beryllium blocks in SVR 20 to collect samples of soil moisture for chemical and radiological analysis. Attempts to draw samples from the 2-m (6.6-ft) -deep lysimeter were unsuccessful because of limited sample recovery. Either the vapor ports were plugged, or the soil surrounding the vapor ports had very low permeability. These ports were abandoned to prepare for the Operable Unit 7-13/14 Early Actions Beryllium Encapsulation Project in 2004. Samples were successfully collected from the 6-m (20-ft) -deep lysimeter during 1997 and 2000. Tests for chloride were made in the sample analysis because magnesium chloride dust suppressant was applied on SDA roadways in 1984 and 1985 and again in 1992 and 1993. Dissolved magnesium chloride in soil pore water would accelerate corrosion of buried beryllium.

Chloride concentration at 6 m was 675 mg L<sup>-1</sup> in 1997, and decreased to 102 mg L<sup>-1</sup> in 2000 (Hull and Bishop 2003). Chloride concentrations measured in soil-moisture samples from various depths collected adjacent to, but outside of the SDA, ranged from 22 to 340 mg L<sup>-1</sup> with a mean of 106 mg L<sup>-1</sup> (Koeppen et al. 2004). Concentrations measured in 1997 were higher than background concentrations outside the SDA, but less than the 2,650-ppm estimate used to derive the beryllium corrosion rate for the RWMC Performance Assessment (Maheras et al. 1994). Elevated chloride concentrations measured around the beryllium blocks are interpreted to be related to use of magnesium chloride dust suppressant on SDA roads. A lack of comparable data from adjacent wells (i.e., Wells W-05 and W-06), and removal of lysimeters in SVR 20 make it difficult to quantify the vertical movement of chloride at this location.

**3.9.2.7 Type B Probes.** Several additional Type B probe vapor ports were installed at SVR 20 in 2001 to augment the three original sampling locations at the GSP-1 borehole (see Figure 3-52). Only Type B Probe SVR20-5-VP3 produced samples. All Type B probes in this area were abandoned to prepare for the Operable Unit 7-13/14 Early Actions Beryllium Encapsulation Project in 2004. Type B Probe SVR20-5-VP3 was approximately 3.5 m (11.5 ft) from the centerline of the buried beryllium with vapor ports at a depth of 5.4 m (17.7 ft). This probe was farther from the buried beryllium than the original monitoring points at GSP-1 and provided useful information about the lateral migration of tritium and C-14 through November 2003.

### 3.9.3 Activated Metal Monitoring

A large fraction of the total SDA inventory of C-14 is present in activated steel. The release rate of C-14 from activated steel, accelerated by corrosion, is being studied at SVR 12 (see Figure 3-52). Because SVR 12 is parallel to the interior perimeter road along the eastern edge of the SDA, it is not likely to be affected by roadway activities (e.g., snow clearing or magnesium chloride dust suppressant). The area is not in an obvious run-off path, and there is little vegetation.

Highly irradiated, activated stainless steel end pieces from spent Experimental Breeder Reactor II fuel elements were buried in SVR 12. Spent fuel elements from Experimental Breeder Reactor II were sent to INTEC for processing after use. The stainless steel end pieces were physically separated from the fuel in underwater basins at INTEC and sent to the SDA in 10 shipments. Personnel familiar with the disposals indicated that the end pieces were placed in open-top cask inserts, which were perforated at the bottom to allow for draining, on removal from the storage basin (Salomon 2001).

Type B probes were installed near SVR 12 in 2001 (see Figure 3-52). Type B probes at SVR 12 include both vapor sampling ports and tensiometers. Vapor ports were installed to provide information that could be used to characterize release and lateral migration of C-14 from the activated steel. Tensiometers were installed to measure soil matric potential near activated metal in SVR 12 to determine hydraulic gradients and the direction and soil-moisture flow rate. Soil-gas samples have been collected quarterly from the SVR 12 Type B probes since November 2001. In general, the specific activity of C-14 in carbon dioxide near the activated steel is at least two orders of magnitude less than the specific activity measured near activated beryllium in SVR 20. Compared to beryllium, activated stainless steel corrodes slowly and is a much less intense source of C-14.

### 3.9.4 Transport Studies

The initial radiological Performance Assessment for the SDA (Maharas et al. 1994) assumed all released C-14 remained as a soluble, unretained component of soil water, potentially migrating downward toward the aquifer. However, because C-14 carbon dioxide can partition into the gaseous, solid, or liquid phases, studies were initiated to improve our understanding of C-14 mobility in the vadose zone and subsequently refine our estimates for risk and exposure. Aqueous transport studies by Hull and

Hohorst (2001) using soil from RWMC indicated that dissolved C-14 was retarded relative to the movement of water and that atmospheric loss of C-14 could result in an expected reduction in radioactive dose in the aquifer from C-14 to be between 5 and 10% under dry conditions. These initial small-scale saturated column studies led to larger near-field-scale studies to describe multiphase transport of C-14 in RWMC soil (Plummer, Hull, and Fox 2004; Fox et al. 2004). The mesoscale column was designed and constructed to model vadose zone conditions in the SDA and allowed researchers to monitor aqueous- and gas-phase transport over relatively long distances over time.

Results after 1 year of operation showed that 66% of the C-14 (added aqueously as C-14 labeled bicarbonate) exited the column as C-14 carbon dioxide gas and that only 3.7% exited the column as effluent. The remainder either was adsorbed in the soil or remained within the pore water and soil gas. Data from column studies were used to calibrate multiphase reactive transport models for C-14 (see Section 5.4.1). The models indicated that C-14 transport in unsaturated soil is described by reactive diffusive transport in the gas phase with little aqueous-phase transport or solid-phase retention. One- and two-dimensional transport models also were developed to investigate simultaneous local release of C-14 and tritium from buried beryllium blocks in SVR 20 (Nalla 2004) (see Section 5.4.2). Models were calibrated with 5 years of monitoring data from SVR 20. In a 22-year simulation, the model predicted that diffusional migration of C-14 carbon dioxide to the atmosphere accounted for 82% of the cumulative C-14 released; cumulative surface migration of tritiated water was less than 1%. Results from this model predict that a large fraction of the released C-14 carbon dioxide is lost to the atmosphere, while most of the tritiated water is retained in the subsurface.

The overall conclusion from these studies is that the simplified assumptions of either 100% loss to the atmosphere, as used for reporting under the National Emission Standards for Hazardous Air Pollutants (Olson et al. 2003), or 100% migration to the aquifer, as used in the initial radiological Performance Assessment (Maheras et al. 1994) and the ABRA (Holdren et al. 2002), were improved in the modeling for this RI/BRA (see Section 5.4 for a description of the modeling and the assumptions regarding gaseous- and aqueous-phase transport). Allowing C-14 to partition from the aqueous-phase into the gaseous-phase and diffuse through land surface provides a more representative and mass-consistent simulation of C-14 transport.

### **3.10 Criticality Safety Study of the Subsurface Disposal Area**

Sentieri and Taylor (2003) prepared a safety study to address the uncertainty of a criticality (i.e., a self-sustaining nuclear reaction) occurring in buried waste in the SDA to support the Operable Unit 7-13/14 comprehensive RI/FS. The study assessed postulated plutonium criticality scenarios by first defining waste configurations that would enhance the probability of a criticality event, then assessing sensitivity of critical configurations to various parameters affecting critical systems. Criticality occurs under an extremely limiting set of circumstances: large masses of fissile material in the appropriate configuration and in the presence of a moderator (i.e., water or some other substance that slows neutrons to enhance fission). Parameters affecting criticality in a fissile system include (1) mass of fissile material present, (2) presence of moderating material, (3) geometric configuration, (4) presence of diluents and neutron poisons, (5) reflection conditions around the system, and (6) concentration and distribution of the fissile material in the waste. Sentieri and Taylor (2003) evaluated each of these parameters and their effect on reactivity.

Buried in the SDA is an approximate cumulative total of 1,100 kg (2,425 lb) of Pu-238, Pu-239, and Pu-240 (plutonium is a fissile isotope: one that fissions more readily than other isotopes), with an approximate combined activity of  $9.9\text{E}+04$  Ci. Most of the plutonium waste in the SDA was received from Rocky Flats Plant. Criticality would not be a concern if the waste was in compliance with fissile material disposal limits that were set at Rocky Flats Plant. The limits were 200 g of fissile material per

drum or 5 g of fissile material per cubic foot, with a total loading not to exceed 350 g per box. However, Stored Waste Examination Pilot Plant assays of stored waste at the TSA indicate that some Rocky Flats Plant drums exceeded disposal limits for plutonium (Ravio et al. 1995).

Three Rocky Flats Plant waste streams were chosen for analysis based on the perception that they could contain enough plutonium to result in a criticality in the SDA. The waste streams were identified based on limited historical Stored Waste Examination Pilot Plant assay data from stored waste at the TSA and engineering judgment. Waste streams included glove box HEPA filters, graphite molds, and magnesium oxide waste. The HEPA filters were chosen for analysis because overloaded drums containing this waste matrix have been discovered in aboveground storage operations at the TSA. In addition, HEPA filters historically have higher fissile loading than other waste. Graphite molds were chosen based on historical data indicating the possibility of high fissile loading and on the characteristic of graphite as being a good neutron-moderating material in large systems of somewhat homogeneous distributions of fissile material. Magnesium oxide also was chosen because overloaded waste drums containing this waste matrix have been discovered in aboveground storage operations at the TSA.

Sludge waste from Rocky Flats Plant was not considered because of the sludge waste forms and the historically low fissile loading in the sludge matrices. Most of the sludge contains a large amount of carbon tetrachloride, which is a very good neutron poison. Chlorine in the carbon tetrachloride is an excellent neutron absorber that effectively lowers reactivity of the system by removing neutrons, thus reducing the likelihood of criticality. In addition, sludge inhibits optimum conditions required to form a critical system.

The analysis considered combinations of dumped containers and stacked containers with mass and geometry defined to enhance the probability of criticality. A more reactive geometry is provided by stacked containers than randomly dumped containers, according to the analysis. Following the analysis of geometry, the amounts of moderation, neutron reflection, and neutron poison were varied to assess the potential for a configuration that could achieve criticality. Extremes of perfect neutron reflection and perfect moderation were assessed. Postulated configurations of the analysis created near-ideal conditions and would be more reactive than the actual waste conditions within the SDA.

None of the configurations analyzed was conducive to spontaneous criticality. In summary, to achieve criticality with minimal mass, several conditions must occur simultaneously:

- The mass of plutonium must be larger than 520 g
- Water must be present in sufficient amounts to start a reaction
- Plutonium must be homogeneously distributed within the fissile volume
- Geometry must be optimal
- Diluting material or neutron poisons cannot be present in the system.

Within the SDA, these optimal conditions for criticality do not occur. Waste is intermixed with neutron poisons (e.g., chlorinated solvents, sludge, and other waste debris). Geometry is not optimal because the plutonium present in the waste is in small, isolated masses. Substantial quantities of water necessary to moderate and sustain a reaction are not present. In addition, metallic plutonium originally contained in the waste when it was buried would not remain in the metallic form but would transform to plutonium oxide in the environment, thus reducing the likelihood of a criticality even further. Soil mixed with the waste would dilute plutonium concentrations, and the soil contains additional neutron poisons.

### 3.11 References

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